Do you think that a centrifugal blower would cause enough pulsation?

HACL Comment: As far as I know, we have some pretty good evidence in certain bag filters with centrifugal blowers, that if you get the blower and distance just right to the bag collector that the bags are vibrating all the time, the dust keeps coming off of them, and they tend to self-clean. So, I would believe that you could get air-column vibrations that would appear certainly at the surface, the back surface of the filter. This is really not steady flow, if you analyze it. It has got the cycle chopping of the centrifugal vane.

Comment by the Speaker: The filters were tested in place, just prior to this 30-day run with the DOP mist test. The DOP test size in this case was about 0.9 microns; and the efficiency of the filters with this test showed about 99.99+. A DF of about .020.

Session Chairman: I should like to thank Messrs. Anderson, Arnett, Green, List and Modrow for a very stimulating number of papers, as evidenced by the questions and discussions.

SESSION VI - SPECIAL PROBLEMS: RARE GASES AND INCINERATORS

Morning - 23 October 1963

A. J. Breslin, HASL, Chairman

Session Chairman: This is Session VI - Special Problems: Rare Gases and Incinerators. The ground rules are as before. I will ask you to hold your questions and discussion period until after the three papers have been presented.

The first paper is entitled, "Use of Foam for Containment and Cleanup of Radioactive Aerosol and Gases," by R. E. Yoder and L. Silverman, HACL, and will be presented by Dr. Silverman.

FOAM SUPPRESSION OF RADIOACTIVE IODINE AND PARTICULATES

Robert E. Yoder

and

Leslie Silverman

Harvard University School of Public Health

INTRODUCTION

The methods for the control of radioactive materials which may escape from nuclear reactors during normal operations or in an accident situation may be grouped into two broad areas. These are: a) containment in which the entire reactor is surrounded by an unfired pressure vessel designed to retain all the materials released from the reactor and, b) confinement in which the enclosed air surrounding the reactor is cleaned by filtration and/or scrubbing to remove any released materials before discharging the air to the environment.

Recent investigations by Silverman et al. (1, 2) have shown that iodine can be rapidly removed from contaminated air spaces by filling the volume with a high expansion foam similar to that used in fire fighting. (High expansion foams have expansion ratios $\sim$1000:1 whereas fire fighting foams have expansion ratios of $\sim$30:1. The expansion ratio is defined as the volume occupied by the foam to the volume of liquid used to produce the foam.) The foams which he used were water solutions of an ether lauryl sulfate surfactant containing a foam stabilizer. By means of standard chemical detection methods it was shown that the time required to reduce the air concentration of normal iodine to 5% of its initial value was one-tenth of that required when no foam was used. He also demonstrated that an aerosol particle concentration was significantly reduced with the addition of foam to the aerosol chamber.

These results prompted Silverman to suggest that a foam generator would make a useful stand-by decontamination system for reactors depending on containment for gaseous radioactivity control. There are several questions which must be answered before foam containment systems can be used in practice. One important question involved the radiation stability of such foams. No data were available as regards the levels of ionizing radiation which might destroy the foam. A second question involves the stability of the foam with live steam which might be present after a water reactor accident. The foam selected must withstand the high temperature and condensing action of the steam and must retain the captured fission gases. Another important aspect is the release of captured gases upon foam collapse by water spray. Not only must the foam remove iodine, but it must also fix the iodine in such a manner that it is not released. The use of sodium thiosulfate in stoichiometric excess as a primary foam additive

was to be investigated to determine the completeness of the sodium thiosulfate-iodine reaction.

After the foam collapse by water sprays, residual foaming agents in the water may cause frothing and foam production at the inlet of liquid storage facilities. Suitable anti-foam agents need to be selected to minimize subsequent foam production.

OBJECTIVES

Arrangements were made late in 1962 to perform a series of radioactive experiments in a 6000 cubic foot hot cell at the Oak Ridge National Laboratory. The chief purpose of these tests was to evaluate the effects of high activity levels, steam, water and general scale up of previous laboratory studies on the efficiency of foam supression systems.

Aside from the problems concerning the fate of radioactive gases in foam environments, problems involving particulate removal and particulate-iodine interactions must be considered. In addition to the iodine tests which were performed, two experiments were performed in which irradiated uranium foils were burned to produce a radioactive aerosol in conjunction with the released iodine. The fume produced by burning the uranium foils was combined with the iodine release.

METHODS

A. Apparatus. Figure 1 is a schematic drawing of a typical foam generator used in this study. A 2.5% solution of foam concentrate in water is sprayed from a nozzle onto a loosely woven nylon mesh fabric. The latter retains a large fraction of the foam solution. The blower, generally an axial flow fan in large units > 10,000 cfm and a centrifugal blower in smaller units, is adjusted to supply sufficient air to create bubbles from the liquid film on the cloth. This foam solution may be gravity fed or pumped to the nozzle. Proportioning pumps may be employed also to blend the water and concentrate. In our experiments, the solution was pre-mixed in 55 gallon drums and pumped to the spray nozzles.

Three foam generators were built for the hot cell experiments - two 1,000 cfm units and one 2,000 cfm unit. Figure 2 shows the placement of the generators in the hot cell. Each device was separately controlled to permit flexibility and to effect a comparison of the units. Placement of air inlets to the fans at the top of the cell maximized air recirculation. Foam generation and air encapsulation took place at the bottom of the cell.

A single furnace was used to volatilize the elemental iodine or to burn the uranium foil as required. The furnace shown in Figure 3 consisted of a stainless steel cavity with a funnel shaped entrance guide at the top. The cavity was surrounded by Cal-rod heaters cast in aluminum, the whole assembly was contained in a stainless steel can. The heater which operated at power levels up to 1500 watts was controlled by a powerstat on the control panel.

Elemental iodine-131 and iodine-127 carrier in a quartz ampoule was placed in a stainless steel container as shown in Figure 4, which could fit into the cavity of the furnace. The ampoule was broken by means of a breaker tool inserted through the top of the hot cell. The uranium foil was placed in a similar unit, the only difference being that several holes were drilled in the bottom to the container to allow oxygen to be blown over the burning foil.

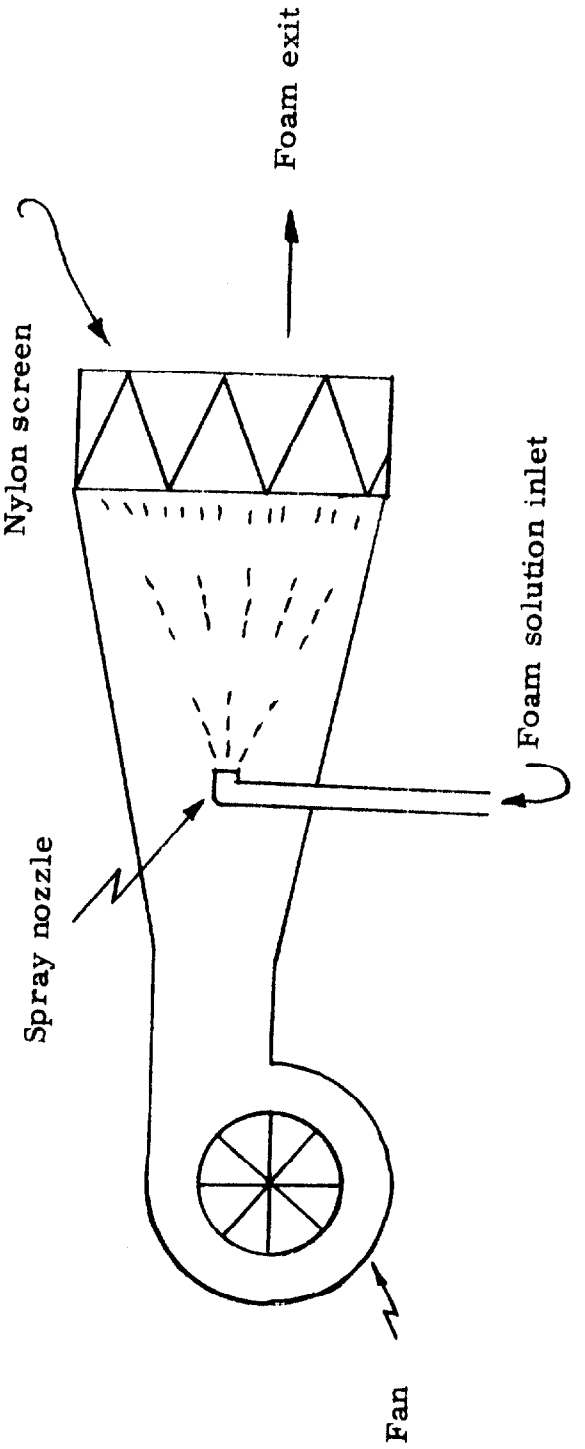
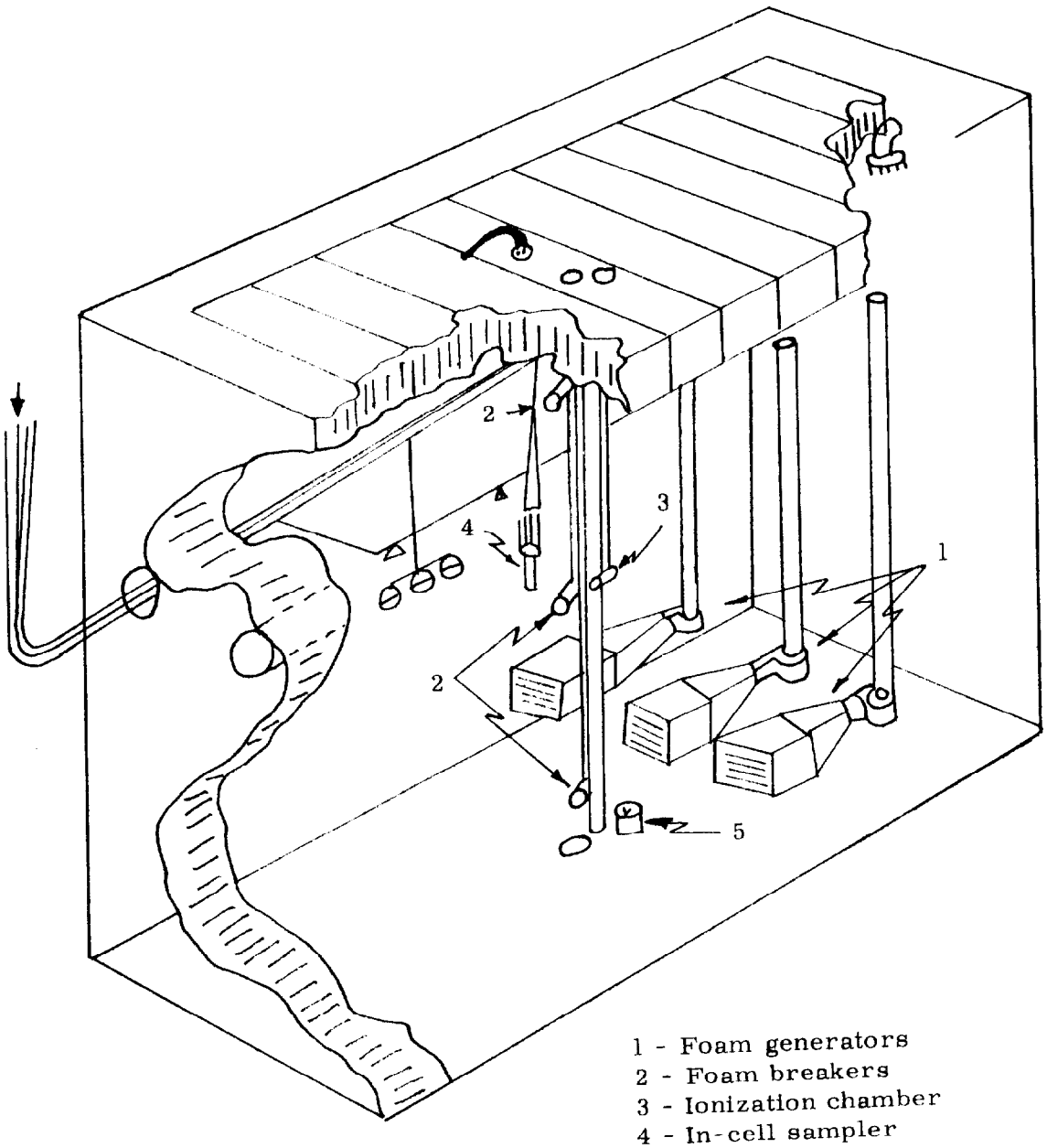


Figure 1 - Schematic Diagram of a Foam Generator



1 - Foam generators
2 - Foam breakers
3 - Ionization chamber
4 - In-cell sampler
5 - Furnace
6 - Sample lines and services

Figure 2 - Schematic Diagram of the ORNL Hot Cell

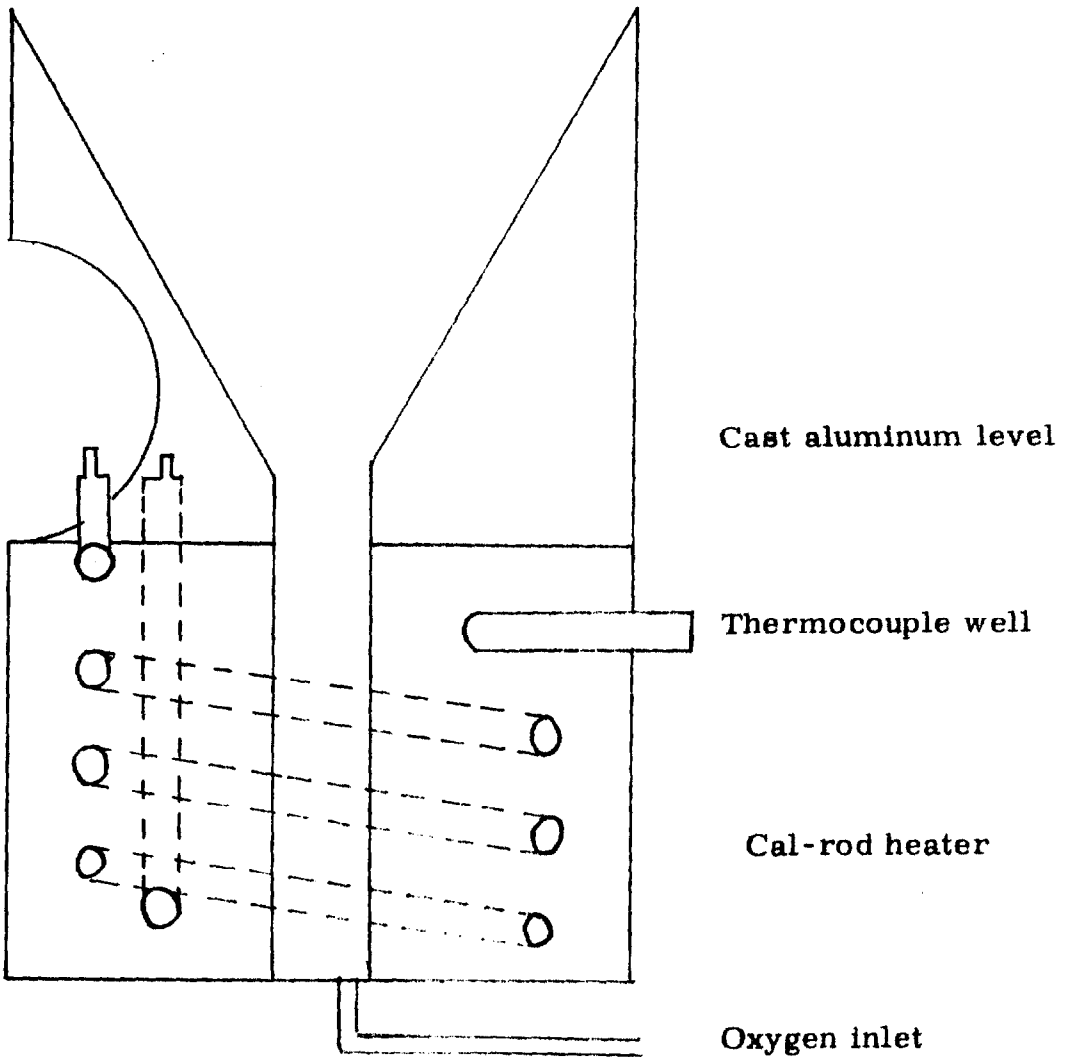


Figure 3 - Schematic Diagram of the Furnace

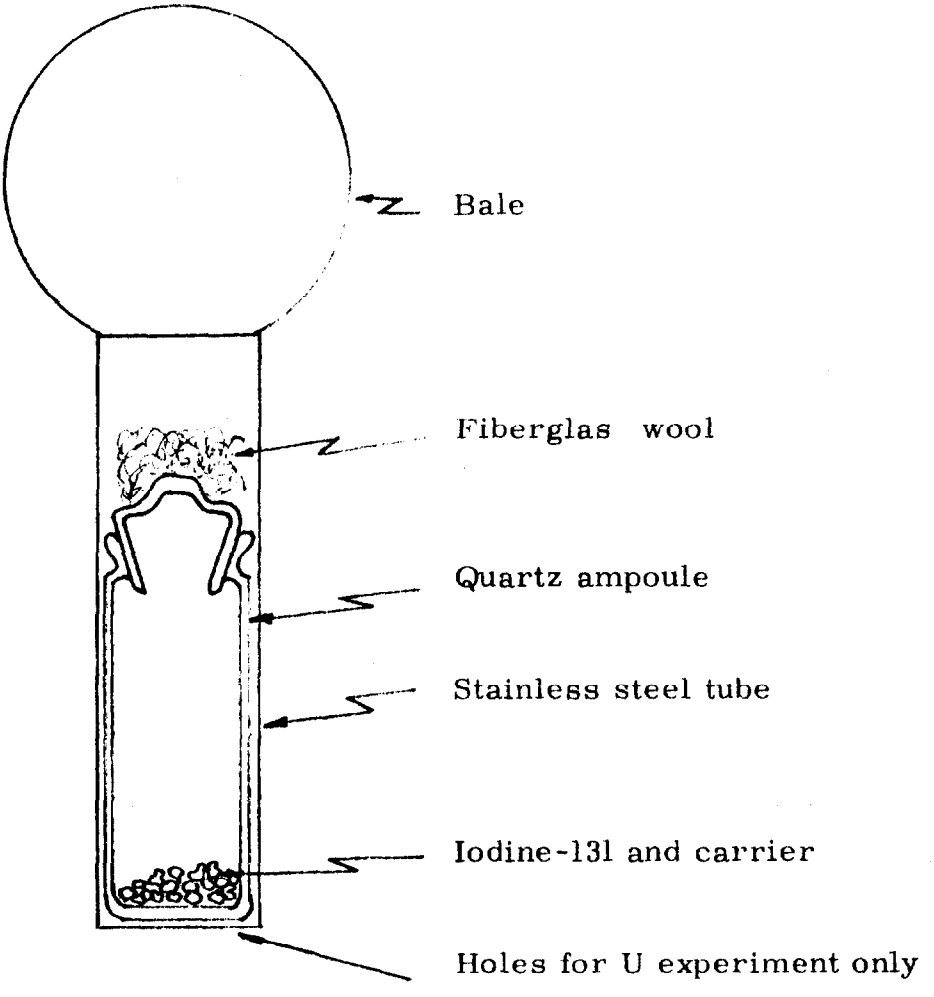


Figure 4 - Iodine Ampoule in the Carrier

The cell atmosphere was sampled at three locations through separate sampling lines as shown in Figure 2. A foam breaker was located at the inlet of the sample lines to prevent any introduction of foam into the lines. The foam breakers were 1½ inch pipe nipples containing an electrically heated helical nichrome wire stretched across the inner diameter. The heat generated by the wire broke the foam, thus releasing the contained gas to the sampling line. The three sampling points were located at different distances from the sample collectors on top of the cell, so each sample line was made to the same length, 50 feet, to minimize variations in wall adsorption. The excess length of tubing was placed in a coil outside the cell face.

Each sampling line was connected to a manifold which held 9 charcoal adsorbent sample collectors. Each sampler was isolated from the manifold by a check valve to prevent accidental contamination of the charcoal by iodine diffusion from the manifold. The sampling lines and manifold were continually purged with air from the cell.

The charcoal samplers were polyethylene drying tubes, 5/8 inch in diameter and 8 inches long, fitted with a hose connection at each end. A small pad of 0.5 micron spun Fiberglas was placed in each end of the tube to support the charcoal, about 17 grams per tube. Figure 5 is a schematic diagram of the sampling manifold and a sampler tube.

In addition to the above sampling system a special sampler was developed which could be inserted directly into the cell. A schematic diagram of one of these devices is shown in Figure 6. Two of these units were fabricated, each holding four charcoal sampler tubes. A single tube with a check valve was lowered into the cell to determine the iodine concentration before the foam introduction. To prevent external contamination, each tube was sealed in a removable cellophane bag which was removed when the samples were withdrawn from the cell.

An ionization chamber was located as near to the center of the cell as possible. This was used to check the initial iodine inventory when the ampoule was inserted into the cell and also to monitor the cell activity during an experiment.

B. Procedures. After each carbon sampler was removed from its sample position it was checked for radioactivity with a Geiger counter and heat sealed to prevent iodine loss. The samples were analyzed for radioactivity either in a high pressure ionization chamber or in a scintillation spectrometer (the data being recorded as microcuries of iodine-131 in each sample). Suitable standards were prepared by the counting laboratory personnel by placing a known quantity of iodine-131 on a series of charcoal samplers. Several samples were observed to determine isotope purity, and there was no evidence of contamination in the iodine tests.

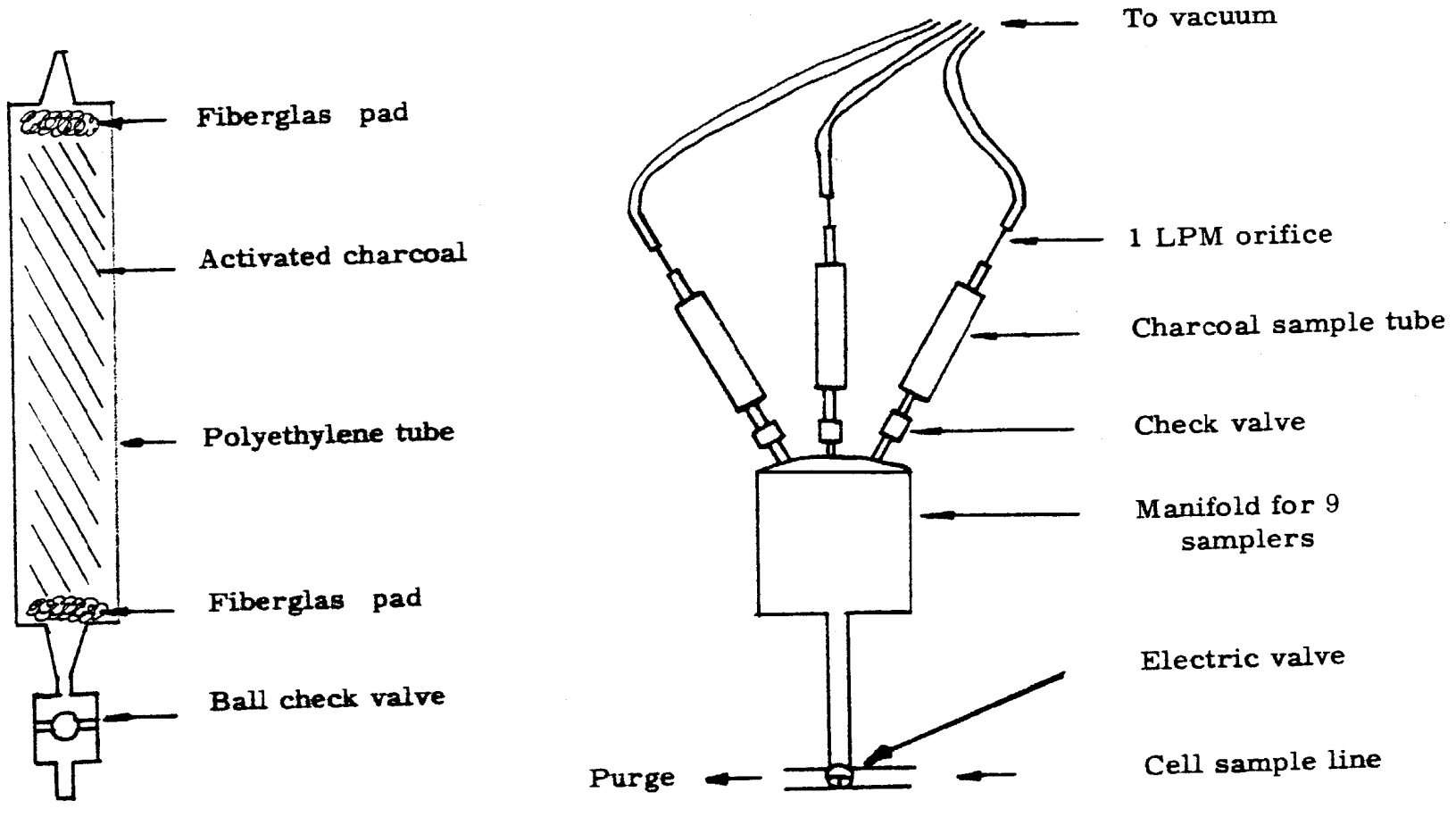
The uranium samples were analyzed in a similar manner. In addition, the samples were examined in a multichannel spectrometer to determine the various radioactive species present.

Water samples of the collapsed foam and spray water were taken after each test and analyzed for activity.

C. Sequence of Experiments. Before sealing the cell, the steam tests were performed, as well as the foam fill time tests. After sealing the cell, a series of tests were performed to measure the air leak rate into the cell.

The five scheduled iodine tests were conducted as follows:

Figure 5 - Schematic Diagram of a Charcoal Sampler and Sampling Manifold



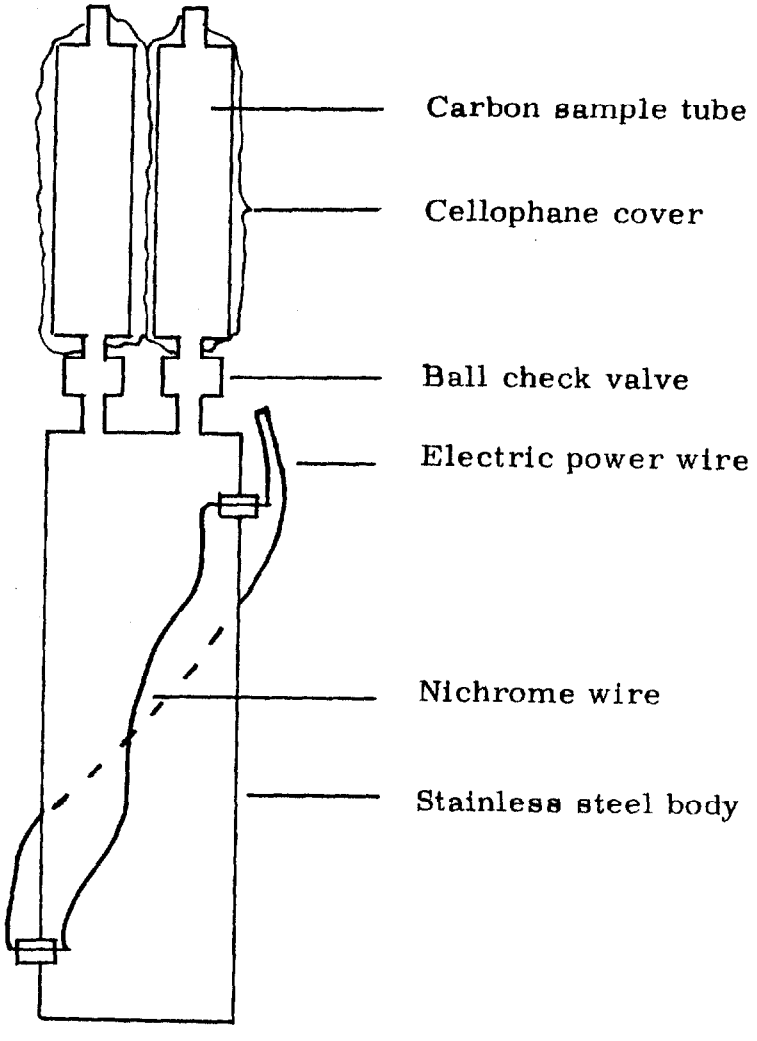


Figure 6 - Schematic Diagram of In-Cell Sampler

1. Iodine was released and its decay measured by cell leakage and diffusion to the walls with the recirculation fans turned on. This test was to provide a baseline for subsequent tests.

2. Iodine was released and then the cell was filled with foam, the foam generators and recirculating fans were then turned off. Two tests were performed in this manner to increase the reliability of the data.

3. Iodine was released and the cell was filled with foam, the foam generator was turned off and the recirculating fan remained on. Two tests were performed using this procedure. Initial data using normal iodine showed that recirculation of the contaminated air tended to improve the iodine capture by foam.

There were two uranium tests in which an irradiated uranium foil was burned and the cell filled with foam. The recirculating fans were operated in the manner showing the best results from the iodine tests.

RESULTS

A. Foam Generator Performance. Table 1 shows the expansion ratios obtained at Harvard and those obtained at Oak Ridge. Part of the poor performance at Oak Ridge was produced by the blower dampers vibrating out of position. This caused an improper air-liquid ratio. The addition of sodium thiosulfate also contributed to a lower expansion ratio. The units were used in various combinations during the several tests because visual observations indicated poor performance. By the end of the series of tests only a single 1,000 cfm unit was operating; the others had developed electrical or adjustment malfunctions. The time required to fill the cell was 10-15 minutes at 2000 cfm.

B. Steam Tests. Steam at 400 pounds per hour was injected into the open cell filled with foam. There seemed to be no deterioration of the foam present. The three foam generators easily replaced any foam collapsed by the injection of the steam and the height of the foam in the cell was raised during the test to a level exceeding that before the injection of the steam.

C. Foam Collapse Test. Water was sprayed into the foam filled cell from 6 spray nozzles located around the periphery of the cell. The total water flow rate was ~27 gallons/minute. The 6,000 cubic foot cell required approximately 250 gallons of foam solution per test (130 to fill and 120 to maintain the foam level over a 2 hour test period). Approximately 1800 gallons of water were required to collapse the foam. Dow Anti-foam A was added to the water according to the manufacturers directions with the result that no subsequent foaming occurred in the disposal of the liquid wastes.

D. Iodine Tests. A reactor operating for 10^8 seconds (approximately 3 years) at a flux of 10^{13} neutrons/cm^2-sec. has an I^{131}/I_{total} ratio of 1/44, with a total iodine inventory of 1250 grams. To stay within these limits of mass concentration and to provide adequate activity for sampling, one gram of I^{127} was added to the 10 curies of I^{131} which contained 100 milligrams of carrier. This provides a ratio much less than 1/44, but was the best compromise available. The surface area of the hot cell was 1.94×10^6 square centimeters and a monomolecular layer of iodine on this surface would require 0.27 grams of iodine. One gram of iodine-127 added to the iodine-131 thus was believed sufficient to preclude the possibility of plate-out of the total activity. The above value of the surface area does not include the area of the fan ducts, nylon screens of the generators or the surface of other equipment.

TABLE 1

FOAM CONCENTRATE EVALUATION AND GENERATOR PERFORMANCE

Generator	Foam Concentrate Designation[1]	Detergent Concentration	Expansion Ratio[2]
18" screen 250 cfm	Blue	1.5%	500:1*HACL[3]
Same as above	Pink	2.5%	1700:1*HACL
Same as above	AY	1.5%	940:1*HACL
Same as above	AX	1.5%	1800:1*HACL
Three generators 4,000 cfm	AY	2.5%	350:1 ORNL[4]
Same as above	AX	2.5%	350:1 ORNL
Two generators 2,000 cfm	AX	2.5%	320:1 ORNL
One generator 2,000 cfm	AX	2.5%	400:1 ORNL

1. Arbitrary manufacturers description. Exact composition not revealed but known to contain ammonium ether lauryl sulfate and lauryl alcohol.

2. Expansion ratio = volume of foam/volume of liquid used.

3. Harvard Air Cleaning Laboratory.

4. Oak Ridge National Laboratory.

* No sodium thiosulfate added.

The only method available to estimate the amount of iodine placed in the furnace was to hold the capsule at the ion chamber level inside the hot cell and read the ionization current. (The capsule at this point was suspended by a string and invariably swinging.) These data are presented in Table 2. The first experiment, 12-19-62, contained 7 curies of I^{131} and the last iodine experiment, 1-7-63, contained 11 curies of I^{131}.*

The release of the iodine from the furnace gas was determined visually by observing the purple vapor evolved and also by noting the reading of the electrometer. After the release the ionization chamber readings increased by a factor of ~10. At the point of maximum reading the samplers were started.

Nine air samples were taken inside the cell and twenty-seven were taken via the heated sample lines - nine through each line. Due to excessive iodine loss in the sample lines useful data could not be obtained from these systems, therefore our data analysis was restricted to the in-cell samples which were considered reliable. All flow rates for sampling with the outside samplers were controlled by critical orifices designed to maintain a flow rate of 1 liter per minute. The in-cell sample flow rates were metered with a Fisher-Porter rotameter.

Since the cell pressure varied as much as + 1" w.g. from the desired pressure of -2" w.g. during the experiments, cell leakage rates also varied by an unknown amount. From orifice theory, based on a coefficient of 0.65, the leakage rate was estimated to be approximately 20 cfm.

The curves presented in Figure 7 show the composite results obtained in the five iodine experiments. "Fan On" means that the air was constantly recirculated in the cell with the foam liquid supply turned off following initial filling with foam. Foam was added as necessary to keep the cell filled. "Fan Off" means that the air was not recirculated (after initial filling with foam) and that the fan was turned on only to generate the required foam volume necessary to keep the cell full. These data show that the foam reduces the iodine concentration above the wall losses by a factor of three, but a comparison with Silverman, et al data (1) indicates that the iodine reduction is not as good as anticipated using chemical detection procedures. The advantage of using foam is clearly demonstrated however. The absolute value of the iodine-131 concentration at the start of the experiments is shown in Table 3 which also shows the final air concentrations, the absolute iodine inventory and the quantity of iodine collected by the foam.

The iodine activity material balance cannot be reconciled completely, as the material was stated to be a nominal 10 curies which was found to vary from 10 to 30 per cent. The iodine concentration was measured at the end of the experiment before and after foam collapse and no significant change in the air concentration was detected. This fact shows that no iodine is released from the collapsed foam.

It is assumed that the unaccounted for activity in the material balance leaked out of the cell or plated out on the inner surfaces of the cell and its contents. The latter assumption is supported by the ionization chamber readings which decrease very slowly after each experiment and which showed increasing initial activity levels for successive experiments. Desorption from the inner surfaces took place because the air leakage into the cell between experiments was sufficient to allow several air volume changes. Figure 8 shows the ionization chamber readings for two comparable experiments. Time zero is the time

* These specific values were supplied by the Isotopes Division, ORNL.

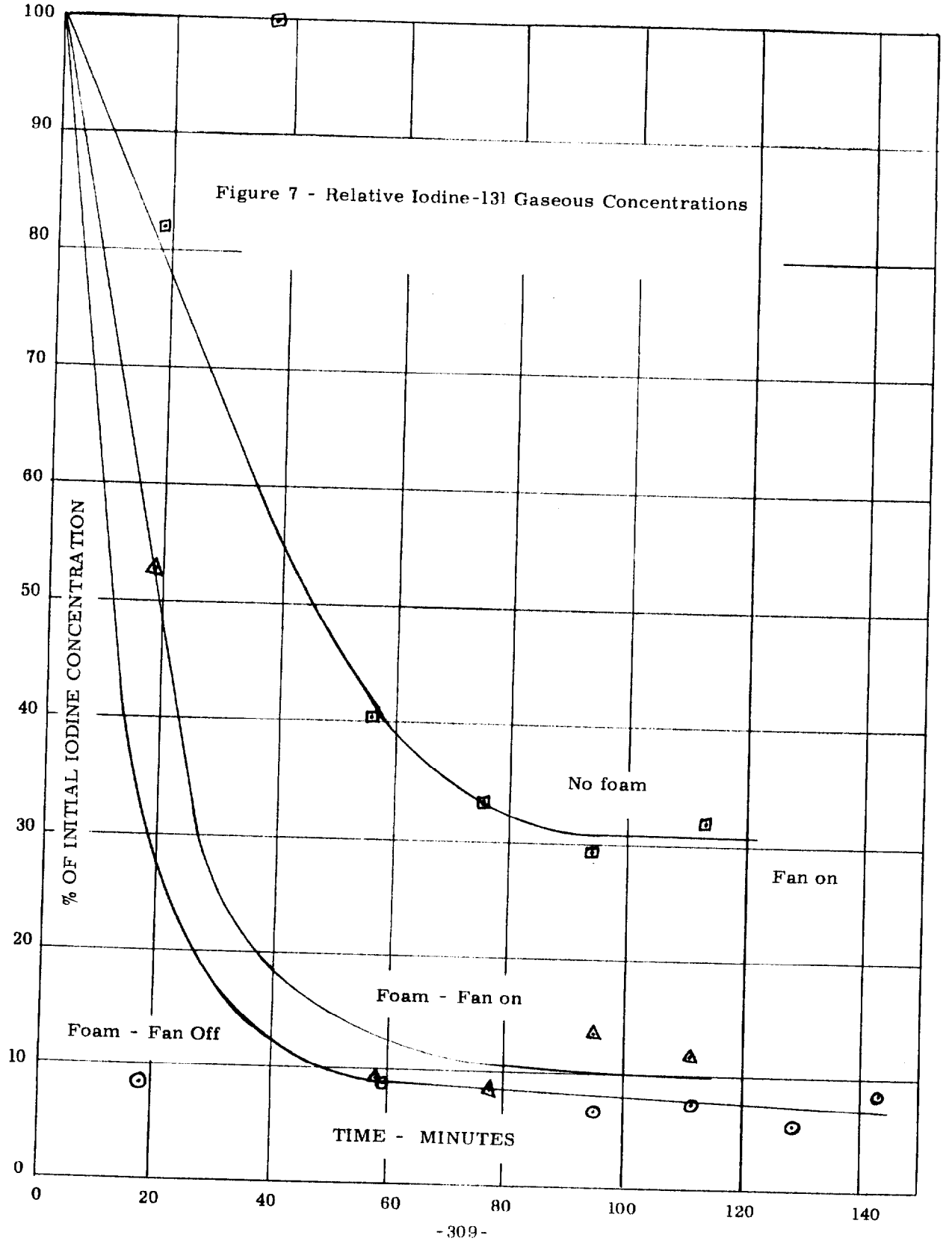


Figure 7 - Relative Iodine-131 Gaseous Concentrations
% OF INITIAL IODINE CONCENTRATION
TIME - MINUTES
No foam
Fan on
Foam - Fan on
Foam - Fan Off
100
90
80
70
60
50
40
30
20
10
0
0
20
40
60
80
100
120
140

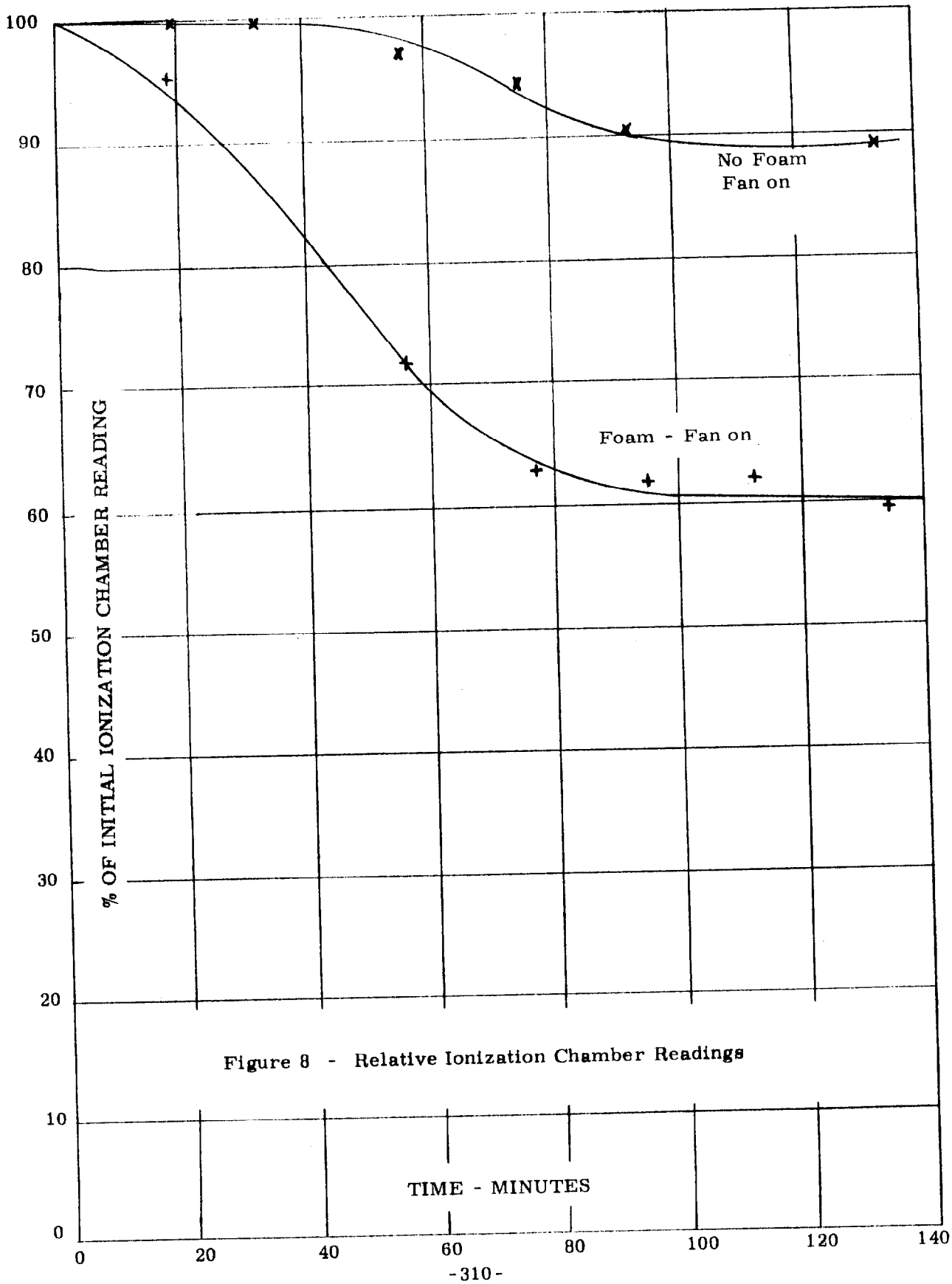


Figure 8 - Relative Ionization Chamber Readings

TABLE 2

IONIZATION CHAMBER READINGS DURING INSERTION OF THE IODINE-131 INTO THE HOT CELL

Test Date	Background Ionization Current (amperes)	Peak Ionization Current (amperes) (ampoule closest to the chamber)	Ionization Current with Sample in the Furnace (amperes)
12-19-62	2.5×10^{-12}	0.8×10^{-10}	8.0×10^{-12}
1-4-63	1.8×10^{-12}	1.1×10^{-10}	3.8×10^{-12}
1-5-63	8.6×10^{-12}	1.8×10^{-10}	1.0×10^{-11}
1-7-63	8.5×10^{-12}	2.6×10^{-10}	1.3×10^{-11}
1-8-63*	1.03×10^{-11}	9.6×10^{-10}	3.4×10^{-11}
1-9-63*	4×10^{-11}	7.4×10^{-10}	5.8×10^{-11}

* Uranium foil burning.

TABLE 3

IODINE AIR CONCENTRATIONS AND MATERIAL BALANCE

Test Date	Test Conditions	Initial Iodine Inventory curies	Iodine in Air Concentration at t = 0 µc/ml	Total curies	Iodine in Air Concentration at end of Experiment µc/ml	Total curies	Iodine Concentration in collapsed foam and spray water total curies	% Iodine Retained in the Cell
1-4-63	No Foam-Fan On	9	8.9×10^{-3}	1.67	2.7×10^{-3}	0.52	0.95	16.3
1-5-63	Foam - Fan On	10	7.0×10^{-3}	1.31	0.7×10^{-3}	0.13	9.1	92.2
1-7-63	Foam - Fan Off	11	7.3×10^{-3}	1.37	0.55×10^{-3}	0.13	9.2	84.6

of the iodine-131 release. The shape of the curves in Figure 8 are similar to those presented in Figure 7 but do not reach the same low value because of the influence of the wall deposited iodine and also the iodine within the foam which has reacted with the sodium thiosulfate.

E. Uranium Tests. Two depleted (0.244% U^{235}) uranium foils were irradiated for 48 hours in the Oak Ridge Research Reactor in a neutron flux of 2.23×10^{14} n/cm^2 - sec. One foil was burned in each experiment. The first foil was allowed to decay for 69½ hours before burning, yielding a total fission product inventory (not counting 50% of the Xe and Kr) of 9.6 curies at the time of the experiment. The second foil was allowed to decay 96½ hours, yielding an activity of 7.9 curies. Approximately 0.34 curies of iodine-131 activity was in the foils.

The two uranium tests were identical, after burning the cell was filled with foam and the recirculating fans turned off since this mode of operation gave the best performance with the iodine-131. Each sample from these tests was observed on a gamma ray spectrometer and only iodine-131 was present, except in two samples in which 0.01% of the gamma activity was from iodine-133. The curves shown in Figure 9 are taken from the in-cell sample data and the activity is reported as iodine. The curve shapes in Figure 9 are dissimilar from those in Figure 7. Only postulations can be given at this time to explain the observed differences.

One explanation is that the iodine is attached to sub-micron uranium dioxide fume particles. Oxygen was blown over the foil during combustion and the oxides could have been ejected at that time. Preliminary tests using an unirradiated foil showed that only UO_2 was formed as a combustion product. If the iodine became attached to the fume particles, then its removal from the air would be much less than with no aerosol present. The diffusion coefficient of a 0.05 micron aerosol particle is 6.9×10^{-6} cm^2 per sec., far less than that for an iodine gas molecule - 0.1 cm^2/sec. Since foam collection is controlled by diffusion, one would expect the aerosol-adsorbed iodine to be more difficult to capture.

In contrast to the iodine tests the ionization chamber readings during the uranium tests did not decrease. Sufficient activity remained within the combustion furnace to completely mask any air concentration changes. For this reason it was difficult to determine precisely the time of activity release. The ionization current remained essentially constant throughout the experiment, at the value shown in Table 2, column 5.

Since time did not permit a test using a uranium foil and no foam, no base line is available for an analysis of the effectiveness of foam removal in these two experiments.

F. Cell Decontamination. After completing the series of experiments the cell was opened and the equipment extracted. The sampling lines were cut into three foot lengths and a four inch section of each length taken for radioactivity analysis. Only iodine-131 was present and the quantities were such that several sections had to be used for a composite sample.

The nylon mesh screens from the foam generators were quite radioactive (5R/hr. at the screen surface 15 days after the last test). This accounts for a large portion of the activity loss from the cell air. No assessment was made as to the composition of this activity.

G. Comparison Between Theory and Experimental Data. Fuchs (3) reports that the reduction in aerosol concentration within a spherical container may

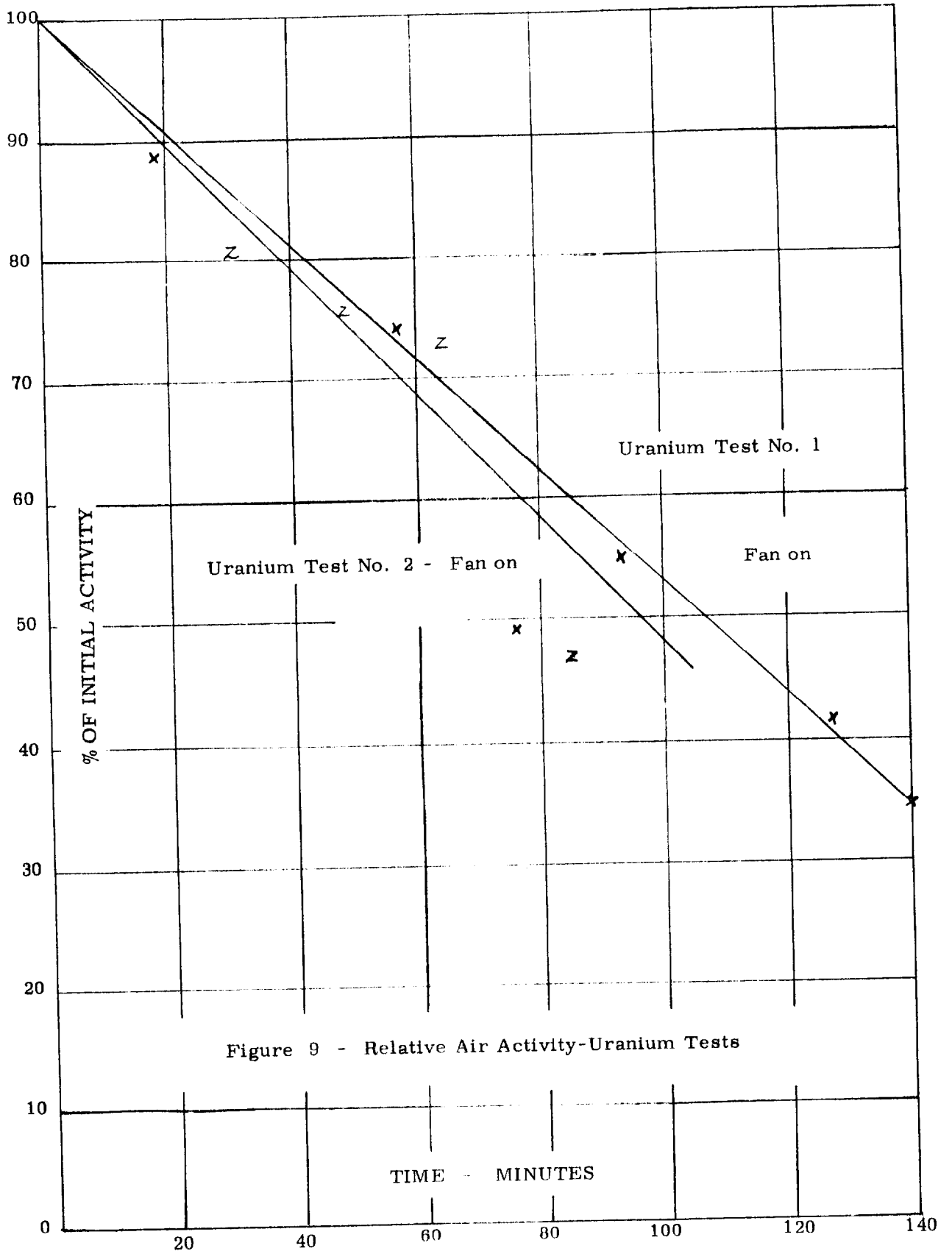


Figure 9 - Relative Air Activity-Uranium Tests

be calculated with the equation:

$$n = n_o \quad \frac{6}{\Pi^2} \sum_{v=1}^{\infty} \quad \frac{1}{v^2} \quad \exp\left(-D \Pi^2 v^2 t/R^2\right)$$

where:

n_o = the initial aerosol concentration in the container

n = the aerosol concentration at any time t

t = time in seconds

D = the diffusion coefficient of the aerosol $cm^2/sec.$

R = the radius of the sphere

v = a whole number

The above equation was developed for particles but for low concentrations of gas the formula should apply with appropriate changes in the diffusion coefficient assuming the gas molecules are readily transferred from the gas phase to the liquid phase and exert no back pressure once in the liquid phase. Sodium thiosulfate was added to the foam solution to meet this requirement as well as to convert the iodine to non-volatile sodium iodide.

Calculations using R = 2.5 cm (approximate size for a foam bubble) and t = 10 seconds predicts a removal of 87.5% of the iodine in this time. This removal rate is greater than that experienced in these experiments, see Figure 7. The size of the bubbles can be estimated only, because of rupture and compression within the foam mass.

At the present time experiments are in progress to measure the rate of iodine removal in single bubbles of controlled size and composition. Preliminary data from these experiments indicate that actual gas transfer rates to liquid absorbing films may be considerably lower than theory predicts. The mass transfer resistance (back pressure) produced by the iodine dissolved in the foam requires a modification of this equation before it can be applied to the data.

CONCLUSIONS

The experiments with radioactive iodine described in this paper confirm previous reports that ether lauryl sulfate foams with sodium thiosulfate reactant can be used to decrease the iodine concentration in a closed chamber. The iodine-131 concentration reduction attributable to leakage, dilution, and wall adsorption was 70% of the initial concentration, 100 minutes after the iodine release. When foam was added to the system, the iodine-131 concentration reduction was 90% with air recirculation and 92% with no air recirculation at 100 minutes respectively.

The rate of iodine concentration reduction was 1.15% of the initial concentration per minute without foam and 2% per minute with recirculation plus foam and 4% per minute with foam and no recirculation. The iodine concentration decreased to a constant value of 30% of the initial concentration in 100 minutes

in the no foam experiment, but required only 60 minutes to reach a constant value of 10% of the initial concentration with foam.

The data shows that the foam used decreased the iodine-131 concentration in a closed cell to a value one-third lower than that measured without foam and the rate of concentration reduction was nearly three times greater with foam than without.

Two uranium foil burning experiments using foam indicate that the presence of particulates alters the pattern of removal which is far less rapid than if no aerosol were present. One important fact observed in these experiments was that very little activity was released from the ignited uranium foils and the only material detected was iodine-131 and 133. The greater part of the activity remained in the furnace as evidenced by ionization chamber readings.

These data do not agree with the theory presented by Fuchs (3) which has been used to calculate the rate of aerosol concentration reduction in a spherical chamber. Further theoretical and experimental work is being carried out to explain the observed discrepancy.

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LIMITS OF SENSITIVITY IN THE MONITORING OF RADIOACTIVE GASES, WITH PARTICULAR REFERENCE TO KRYPTON 85

by

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INTRODUCTION

The detection of radioactive gases is one of the important monitoring functions required around reactor installations. A stack gas monitor is needed to insure that levels of radioactive contaminants will not be released to the environment in quantities in excess of radiation protection guides. Radioactive gas or particle monitors are also often sensitive enough to give indications of possible malfunctions which release activities at levels well below the airborne activity concentrations which could result in excessive releases to the environment.

If we confine ourselves to the consideration of radioactivity in the gaseous phase, thus excluding the detection of radioactive particulate matter, we find that two main approaches are used in practical monitoring systems. One is to collect the gas in an ionization chamber and measure the current produced by its radioactivity. The second is to immerse a Geiger-Mueller or other counter into the gas and count the pulses produced in the detector by the rays emitted by the gas.

Both the ionization chamber and particle counter methods represent gross radioactivity measurements. Their advantage is that they use relatively simple and reliable instrumentation. They cannot be used to any appreciable degree to analyze the radiations and identify the radioactive contaminants, except perhaps by what can be learned from half-life or simple absorption measurements. Other methods, in particular, scintillation methods, must be used if more detailed data are needed.

In this paper we shall present calculations of the efficiencies of idealized ionization chambers and particle detectors. The results will give us an insight into the factors which effect the sensitivities of the detectors and indicate how the sensitivities might be best improved in particular instances. Calculations of the theoretical sensitivities of some common gas detectors will be made and compared with actual measurements.

All fission gases are both beta and gamma emitters. We shall confine ourselves to detection of the beta radiation since in most instances, the beta radiation is the more sensitive indicator of radioactive gas contamination.

The calculations and measurements will deal with Krypton 85 as the radioactive gas. This gas is important as the most potentially serious atmospheric contaminant from the long-term use of nuclear power. Krypton 85 is also being used in medical research as a tracer in studies of the circulation, and must be monitored in connection with this work.

EFFICIENCY OF DETECTION OF SPHERICAL AND CYLINDRICAL IONIZATION CHAMBERS

Consider the current produced in a spherical ionization chamber which contains a radioactive gas. If we increase the size of the chamber, keeping the total amount of radioactive gas in the chamber constant, the fraction of energy emitted by the gas which is absorbed in the chamber will increase. The current produced by the gas will also increase with size and then will level off as the dimensions of the chamber significantly exceed the effective range of radiations within the chamber. The current contributed by the radiation background will increase steadily with increased chamber volume.

An approximation of the fraction of energy absorbed within a spherical ionization chamber may be obtained by considering the absorption of beta particles as exponential. Exponential absorption holds over most of the range of beta particles, with an absorption coefficient by

$$\frac{\mu}{\rho} = \frac{17}{E_m^{1.14}}$$

where μ / ρ is the absorption coefficient in cm^2/gm, and E_m is the maximum energy in Mev. (2)

Equations for the flux from a source uniformly distributed inside a spherical medium are given in Ref. (3). The equations for a sphere of radius R_o, and points P_1 at the center of the sphere, P_2 at the surface, and P_3 within the sphere and at a distance d from the center are:

at P_1

$$\phi = \frac{Sv}{\mu_s} (1-e^{-\mu_s R_o})$$

at P_2

$$\phi = \frac{Sv}{2\mu_s} \left\{ (1 - \frac{1}{2\mu_s R_o} + \frac{e^{-2\mu_s R_o}}{2\mu_s R_o} \right.$$

at P_3

$$\phi = \frac{Sv}{\mu_s} \left\{ (1 - \frac{e^{-b4}}{2} - \frac{e^{-b5}}{2} - \frac{1}{4\mu_s d} \left[e^{-b4}(1+b_4) - e^{-b5}(1+b_5) \right] \right.$$

$$\left. + \frac{b_{4b5}}{4\mu_s d} \left[E_i(b_4) - E_i(b_5) \right] \right.$$

where

$$b_4 \equiv \mu_s (R_o - d) \text{ and } b_5 \equiv \mu_s (R_o + d)$$

μ_s is the absorption coefficient for the radiation, S_v is the number of particles emitted per second per unit volume.

These equations enable evaluation of the ratio of the average dose rate in a sphere to the dose rate in an infinite medium. This ratio, increased by backscattering from the walls of the chamber, is equal to the efficiency of the spherical detector.

The fraction of the total energy emanating from the active volume of a cylindrical chamber which is absorbed within the active volume is given in reference (4) as:

$$F = \frac{E}{3R}\left[\frac{\mathcal{Z}}{2} + 2\left\{\frac{2a(r-a)^2}{2} + \frac{4}{3}r^3\left[\cos\phi\,(\sin^2\phi + 2)\right]_\theta^0}{\pi\,(r^2 - a^2)}\right\}\right]$$

F = fraction of energy emitted in active volume that is absorbed in that volume

E = total energy emitted by radionuclides within volume

R = range (inches of air) of β particles at one-third of maximum energy

$\mathcal{Z}$ = axial length of the active volume of the chamber

r = outer radius of the active volume of the chamber

a = inner radius of the active volume of the chamber (outer radius of the central electrode)

θ = arc cos a/r

ION CHAMBERS FOR RADIOACTIVE GAS DETECTION

Laboratory instruments for measurement of radioactive gases have been described by Tolbert (5). The gas is collected in a specially designed vessel. Particular attention is paid to the insulators and electrodes to minimize leakage current and contact potential. The current is measured by a sensitive electrometer. For greatest sensitivities the vibrating reed electrometer is used. The most sensitive ranges on commercially available electrometers vary between one and three millivolts.

A commercially available spherical ion chamber which we have used in our laboratory has a volume of 2.8 liters. Large chambers, cylindrical in volume, have been used for field monitoring and are patterned after a design by Kanne. These chambers vary between 20 and 50 liters in size and have been discussed in several papers. (4,6)

Using the equations in the previous section, and Krypton 85 as the radioactive gas, we obtain efficiencies of 16 per cent neglecting backscatter and about 20 per cent including backscatter from the chamber walls for the 2.8 liter spherical chamber. For the 20 liter Kanne chamber, the efficiency is 25 per cent. (Krypton 85 data used in the calculations were E_{max} = 0.695 Mev., E_{avg} = 0.23 Mev, u/ρ = 25.8 cm^2/gm, u = .0332 cm^{-1} in air at STP).

The background current produced per unit volume in a field of 0.01 mr per hour would be approximately 10^{-18} amp/cc. This calculation neglects the

contribution of the alpha contamination. If we assume a contamination level of 3 alpha particles per hour per cm^2, use 5.9 Mev as the average energy per particle for radium series alpha particles, and assume that all the energy is dissipated in the chamber, we obtain a background current of 2.1×10^{-19} amperes per sq. cm. of surface. For the 2.8 liter ion chamber with a surface area of 9500 sq. cm. the alpha background is 2×10^{-15} amperes.

Actually, this result is an overestimate since it neglects absorption in the walls. In addition, the alpha contribution can be reduced by keeping the collection voltage low enough to give appreciable recombination in the alpha particle tracks, but high enough to maintain collection of all the ions from beta particles. It will also be shown later that the alpha pulses can be distinguished from the current produced by the beta particles and much of their effect can be excluded in a current reading.

Greatly increased sensitivites can be obtained by compressing the gas into an ionization chamber and taking a current measurement at a high pressure. Working at high pressures, say 50 atmospheres, we obtain not only an increased concentration of gas in the chamber, but a higher fraction of energy absorbed within the chamber. The current from the radiation background also goes up, but apparently not nearly as much as the current from the radioactive gas. Spiers (7) reported an increase of the background response of only a factor of four in raising the pressure of an ionization chamber to 78 atmospheres. The response to gamma radiation increased twenty times.

A modification of a portable ionization chamber discussed by Rising may be useful for monitoring in the field. (8) This ion chamber uses a 30V battery for the ion collection and the battery is mounted inside the chamber between the inner electrode and the connector. This arrangement appreciably reduces leakage current across the insulators and the low voltage results in reduced alpha particle background because of recombination along the alpha track.

SENSITIVITIES OBTAINABLE WITH IONIZATION CHAMBERS

The maximum sensitivity of a radiation detector is determined by many factors, including inherent noise in the system, background radiation, variations in radiation background, the time available to make a measurement, and the desired accuracy of the measurement. Criteria for practical limits of sensitivity for field use cannot be set to cover all applications and conditions and the limits are best established by studying the performance of the equipment under field conditions. As a means of comparing the sensitivities of detectors, it seems reasonable to set the lower limit of detection of the ionization chambers at that level of gas concentration which increases the background current by a factor of two.

If we apply this condition to the 2.8 liter ionization chamber at atmospheric pressure, we obtain a sensitivity of 2.5×10^{-7} μc/cc as the lower limit of detection for Kr^{85}. This is the level which will produce 2×10^{-18} amp/cc in the chamber, the same current produced by a background level of .02 mr/hr, if the chamber is considered to have an efficiency of 20 per cent for the radioactive gas. Lower concentrations may be detected using a 20 liter chamber at high pressure. A decrease of a factor of four is obtained from increased efficiency of detection, and the limit is even lower, dependent on how much the background decreases relative to the response to the source.

Further increases in sensitivity could be obtained by additional shielding around the counter to reduce the background contribution.

The direct measurement of current is limited by the stable resistance available and the maximum sensitivity of the voltmeter. These are 10^{12} ohm and 1 millivolt respectively, although 1 mv measurements require some care in providing proper shielding and minimizing noise signals. Although this indicates a sensitivity of detection of currents less than 10^{-15} amp, it may be preferable to use the rate of charge method for currents less than 10^{-13} amp. In this method, the charge collected by the electrometer over a period of time is measured rather than the "instantaneous" current.

The rate of charge method may, at low radioactivity levels, require several minutes to obtain a measurement. This does not appear to be an important consideration in monitoring at the extremely low levels possible with the method. The sensitivity of the rate of charge method appears limited only by the radiation background. We shall assume that we can monitor practically changes of the order of 50 per cent in the background level and that the lower limits of sensitivity given above can be reduced by a factor of 2 for the rate of charge method.

ANALYSIS OF DETECTION OF BETA RADIATION BY A PARTICLE DETECTOR

We shall now consider the general case of detection of radioactive gases by a particle detector which is immersed in the gas. Assume the detector has unit cross sectional area and the gas is sampled in a medium of dimensions much larger than the maximum range of the beta particles. There will be some attenuation in the detector wall. We shall assume that the attenuation of the beta particles in the medium is exponential over the range and can be quantitatively expressed by an absorption coefficient, μ.

To get the number of beta particles crossing unit area of a detector immersed in the radioactive gas, we have to integrate an expression of the form:

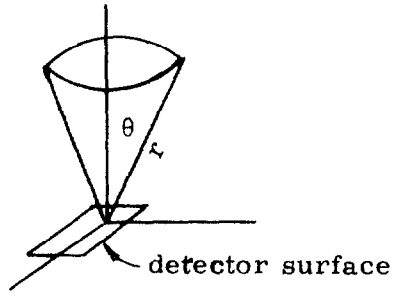
$$J = \frac{No\,\Delta V}{4\pi r^2} \qquad e^{\frac{-\mu\rho r}{\rho}} \cos\theta \; A_w$$

No = the number of beta particles emitted per cc

θ = the angle between the normal to the surface of the detector and the radius vector between the detector and the element of volume ΔV

A_w = an average wall attenuation factor

ρ = the density



$$J = \frac{NoAw}{2} \iint \sin\theta \cos\theta \; e^{\frac{-\mu\rho}{\rho}} \; r \; d\theta dr$$

$$= \frac{NoAw}{4\mu} \left(1 - e^{-\mu r}\right)$$

It is seen that the effect of the radioactive gas at distances greater than a few times ($1/\mu$) becomes small. The number of beta particles crossing unit area in the limit is given by

$$\frac{NoAw}{4\mu}$$

For Kr 85, $\mu = .0332$ cm^{-1} at atmospheric pressure, and $1/\mu = 30$ cm. A volume with dimensions of the order of 1 meter acts essentially as an infinite volume for detection of the beta particles.

A concentration of 10^{-7} μc/cc of Kr 85 gives 0.22 dpm/cc for N_o. Then $J = \frac{.22}{4(.033)} = 1.7$ beta particles/cm^2 -min.

COUNTERS FOR BETA PARTICLE DETECTION

Many types of counters are available for monitoring of radioactive gases - cylindrical, end-window and "pancake" G-M tubes, large-area proportional flow counters, scintillation detectors with thin beta-detecting crystals, and solid state detectors. The choice of one or another of these counters may be indicated by the requirements of a given application.

The Anton 112 cylindrical G-M tube is an inexpensive detector commonly used in radiation survey and prospecting instruments. It is 0.75" in diameter and 3" in length, with a surface area of approximately 45 cm^2. It has a steel wall, 30 mg/cm^2 thick. We shall assume an average wall thickness which is greater than the actual wall thickness by 20 per cent to account for angular variations in the path of the incident beta particles through the wall. The factor A_w for a wall thickness of 30 mg/cm^2 is calculated to be 0.4 for Kr 85. We may thus expect a counting rate for this tube of .4 x 45 x 1.7 or 30 counts per minute at 10^{-7} μc/cc Kr 85.

Mica end-window G-M tubes also make suitable gas detectors. They may be obtained with very thin windows of the order of 1 mg/cm^2, which will produce only a small attenuation of the incident radiation. The thinnest windows are necessary only for the low energy beta emitters such as C-14, but would not be of particular advantage for Kr-85.

The Anton 1008 tube is a double ended mica window halogen quenched pancake tube. The internal geometry is such that its volume and cathode surfaces are considerably smaller than that of a conventional tube having an equivalent mica window surface. Accordingly, its background is low for its detection area. This tube has a diameter of 1.75 in. and a thin window (available from 1.4-4 mg/cm^2). The tube is more efficient than the Anton 112, but is also much more expensive, and less readily available since the manufacturer has recently had production difficulties.

Thin walled proportional flow-counters are obtainable with large detection areas to give a relatively high counting rate for a given gas concentration.

Solid state detectors may be advantageous in gas detection because of the very low attenuation and the small response to background. If we neglect wall attenuation, we estimate about 1.7 cpm/cm^2 area of a solid state detector for a Kr 85 concentration of 10^{-7} μc/cm^3.

Scintillation detectors, employing thin scintillators, may also be used. The thinner the scintillator, the better the beta response relative to the background gamma radiation. Scintillation detectors for beta monitoring are

discussed in reference 9.

If the gas volume viewed by the detector is less than needed for maximum response, increased sensitivity can be obtained by pressurizing the gas. As long as the detector can see a source of Kr-85 beta radiation at least a meter in depth, there is no advantage to compressing the gas to obtain increased counts. However, by compressing the gas to, say, 10 atmospheres, one needs only a 10 cm depth for viewing. This smaller volume is much easier to shield, and facilitates the use of an anti-coincidence system for minimization of counts from cosmic rays. However, the types of detectors which can be used at elevated pressures are limited. The most suitable appears to be the solid state detector.

To complete our discussion of particle detectors, we may also mention internal gas counting methods, i.e. collecting the gas in a counting chamber and counting by some pulse counting method. Shielding plus anti-coincidence methods for reduction of cosmic-ray background can reduce the background to very low levels, and by inserting the gas into the counter, maximum efficiency is obtained.

The minimum size pulse which can be detected is governed by the noise level of the amplifier. Because of the small pulse produced in an ionization chamber by a beta particle, gas multiplication would be required to exceed the noise level sufficiently for detection.

Gas multiplication requires careful gas purification techniques, particularly with regard to the removal of electronegative impurities. For this reason, internal gas proportional counting is not considered an appropriate method for routine monitoring although it is widely used in research work. A high pressure proportional counter is described in reference 10.

SENSITIVITIES OBTAINABLE WITH PARTICLE DETECTORS

The number of beta particles incident per square centimeter per minute on the surface of a detector immersed in Kr-85 was calculated to be 1.7 for 10^{-7} μc/cc concentration. To evaluate the lower limit of detection for any given system, we shall stipulate that the lowest concentration the detector can monitor is that concentration which will increase the background by 50 per cent.

The Anton 112 cylindrical G-M tube has a background of approximately 30 cpm at 0.02 mr/hr. Its calculated response to Kr-85 is 3 cpm for 10^{-8} μc/cc. A level of 5×10^{-8} μc/cc would be required to increase the background by 50 per cent. To detect this activity at the 99 per cent confidence level would require an accumulated count given by $2.6 \frac{\sqrt{n}}{n} = 0.5$, or $n = 27$ counts. One minute counting time would be required for this level of detection.

The background of 30 cpm was measured in the basement of a masonry building and the detector was thus partly shielded. Additional lead shielding did little to reduce the background. Cosmic-ray guard counters in anti-coincidence with the detector would have to be added to give further significant decreases in background.

If the 30 mg/cm^2 wall of the Anton 112 provides excessive attenuation of the incident beta radiation, better performance will be obtained with thinner walled detectors previously mentioned. For Kr-85 beta rays, the very thin walled detectors should give a response about 2.5 times that of the 30 mg/cm^2 wall, per unit detector area. Additional data on the background response is necessary to evaluate the merits of a specific detector. The relative contribution of the background depends on the fraction of the total detector surface which is not used to admit the beta particles into the counter.

MEASUREMENTS WITH G-M COUNTERS AND IONIZATION CHAMBERS ON AIR CONTAINING KRYPTON 85

Measurements were made on air containing Krypton 85 with two commercially available G-M detectors and a 2.8 liter ionization chamber. A known amount of Krypton 85 was injected with a syringe into a 400 cubic foot tank. The krypton had been assayed in the syringe with a 2" by 2" NaI crystal and multichannel analyzer. An Anton 112 cylindrical G-M counter was mounted in the center of the tank and gave the response of the detector in what was essentially an infinite volume of air. The air from the tank was recycled through a 2.7 liter ionization chamber and a 55 gallon drum containing another Anton 112 tube in the center and an Amperex 200 HB end-window G-M tube with a 1.4 mg/cm^2 mica window. The end-window tube viewed the gas from the side of the drum. The arrangement of detectors is shown in Figure 1.

The responses of the cylindrical G-M counter and the ion chamber to a known dose rate from a cobalt-60 source were determined. The radiation background in the laboratory was also measured with the detectors.

The readings are given in Table 1. Column (1) gives the measurements on the Kr-85. Column (2) gives the background when no Krypton was present. This background was approximately 0.02 mr/hr. In Column (3) the background is subtracted from the total reading and the results extrapolated to 10^{-7} μc/cc. In Column (4), the ratio is given of the net reading at 10^{-7} μc/cc to the background.

TABLE 1

READINGS OF 2.8 LITER IONIZATION CHAMBER AND G-M DETECTORS IN AIR CONTAINING Kr-85

Detector	(1) Kr-85 conc.,* 2.18×10^{-6} μc/cc	(2) Background 0.02 mr/hr	(3) Net Reading at 10^{-7} μc/cc	(4) Ratio (3)/(2)
2.8 liter ion chamber	3.6×10^{-14} amp	5×10^{-15} amp	1.43×10^{-15} amp	0.29
Cylindrical G-M counter in large tank	359 c/m	30 c/m	15 c/m	0.50
Cylindrical G-M counter in 55 gallon drum	164 c/m	30 c/m	5.7 c/m	0.19
End window G-M counter in 55 gallon drum	198 c/m	36 c/m	7.4 c/m	0.21

* This value must be considered as tentative, pending calibration with an NBS Kr-85 standard.

The calculated response for the 2.8 liter ion chamber at 10^{-7} μc/cc (efficiency of 20 per cent) was 2.2×10^{-15} amperes, compared to a measured response of 1.43×10^{-15}. The calculated response for the cylindrical counter in the large tank was 30 counts per minute compared to the measured response of 29 counts per minute. The agreement between calculated and measured values is close enough to justify using the calculations to obtain general conclusions. Because in any given situation, a detector will be calibrated with known amounts of Krypton 85, rather than used with a calculated efficiency factor, no detailed investigation into the magnitudes of possible sources of error was made.

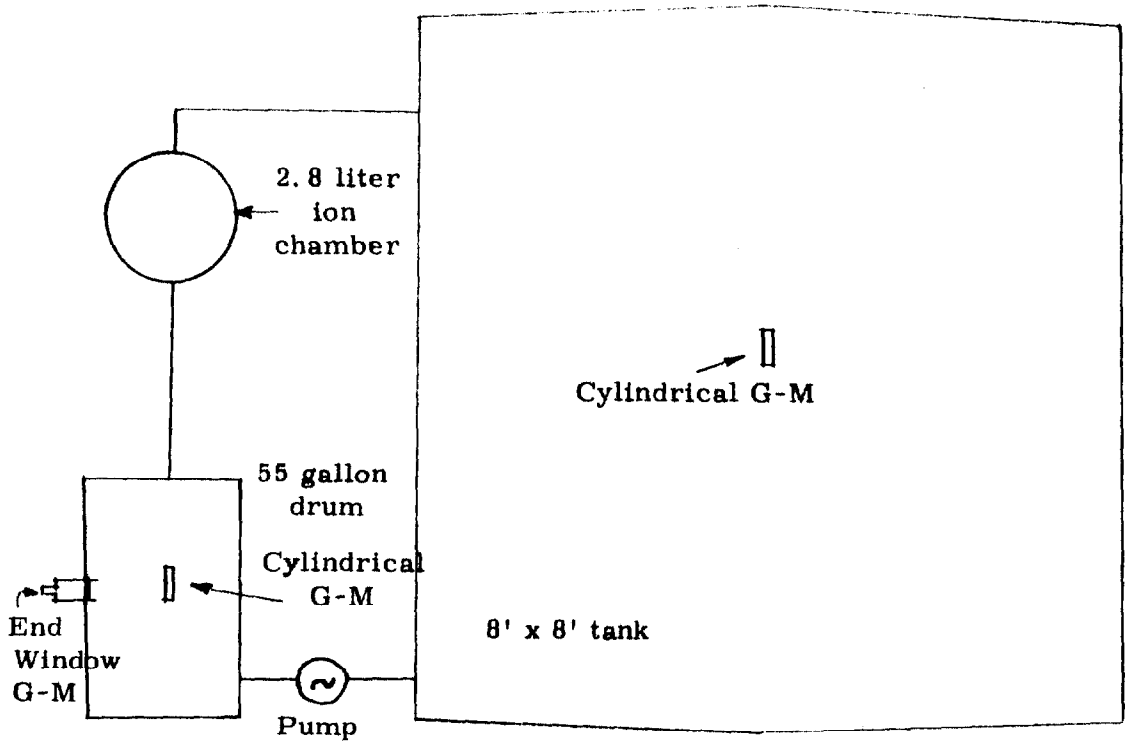


Figure 1 - Arrangement of detectors exposed to Drypton 85

The current in the 2.8 liter ion chamber was recorded on a Bristol recorder. A reproduction of a trace is given in Fig. 2. The trace shows the response of the measuring system when air containing Kr-85 was drawn through the chamber at a sampling rate of 0.2 cfm. It is seen that the recorder responded rapidly to a change in concentration from background levels to 2.2×10^{-6} μc/cc, reaching 90 per cent of its final level in a minute. This illustrates the advantage of a small collection volume for gas sampling. Much larger sampling rates would be needed to get as rapid a response for the detectors in the 55 gallon drum.

The fluctuations in the recorder trace around the mean value are due to the randomness of radioactive disintegrations. The reading of the meter at any time represents an integration of charge over a time interval of 2 RC preceding the instant of measurement, where RC is the time constant of the metering circuit. (11) The current reading will fluctuate according to the statistics of the counts accumulated during the 2 RC period.

The spikes superimposed on the average reading on the trace are produced by alpha particles. At the low current readings they may be neglected easily.

Sensitivities of particle detectors for radioactive gases have been reported in the literature. A G-M detector is used to measure the A-41 concentration of the effluent gas in the stack at the Washington State University Reactor (12). This method was selected after a review of all types of radioactive gas detection systems. The detector gave 10 counts per minute above a 67 cpm background at the maximum permissible concentration.

Ray (13) has used an Eberline Instrument Corporation Large Area flat plate gas proportional probe with a 0.85 mg/cm^2 aluminized mylar window to monitor argon-41 at General Atomics. He reported a net cpm sensitivity of 275 for a stack gas sample concentration of 10^{-7} μc/cc and a 1300 cpm background.

EXPERIENCE WITH IONIZATION CHAMBER MONITOR AT YANKEE ATOMIC ELECTRIC

A 500 ml spherical ionization chamber was installed in the stack gas sampling system at the Yankee Reactor at Rowe, Massachusetts for a period of approximately one month. The current from the chamber was read with a vibrating reed electrometer.

The ionization chamber was located in a 5 mr/hr radiation field which was reduced by shielding to a value which produced a 3×10^{-15} ampere ion current in the chamber. Radioactive gases (Xenons and Kryptons) were released daily to the stack for a short period (5-15 minutes) during sampling of the primary coolant water. The ion chamber - electrometer system responded to each gaseous release by showing an increased ion current at least twice background. The background ion current was $\sim 3 \times 10^{-15}$ amperes and during release the current rose to 10 to 12 $\times 10^{-15}$ amperes depending upon the quantity of material discharged. The 500 ml chamber gave about 0.4×10^{-15} amperes at 10^{-7} μc/cc so the maximum concentration released was 3×10^{-6} μc/cc.

Immediately following an activity release the ion current resumed its initial value prior to release. The lowest stack background recorded was 2×10^{-15} amperes and the highest value recorded was 6×10^{-15} amperes. A one hour room air background was taken each 8 hours. The room air reading was always about $\sim 0.4 \times 10^{-15}$ amperes less than the stack background. This lower value was constant regardless of the level of the stack activity. The reason for the difference has not been investigated. The baseline drift of the electrometer appeared to be sufficiently stable to allow its use as a field monitor.

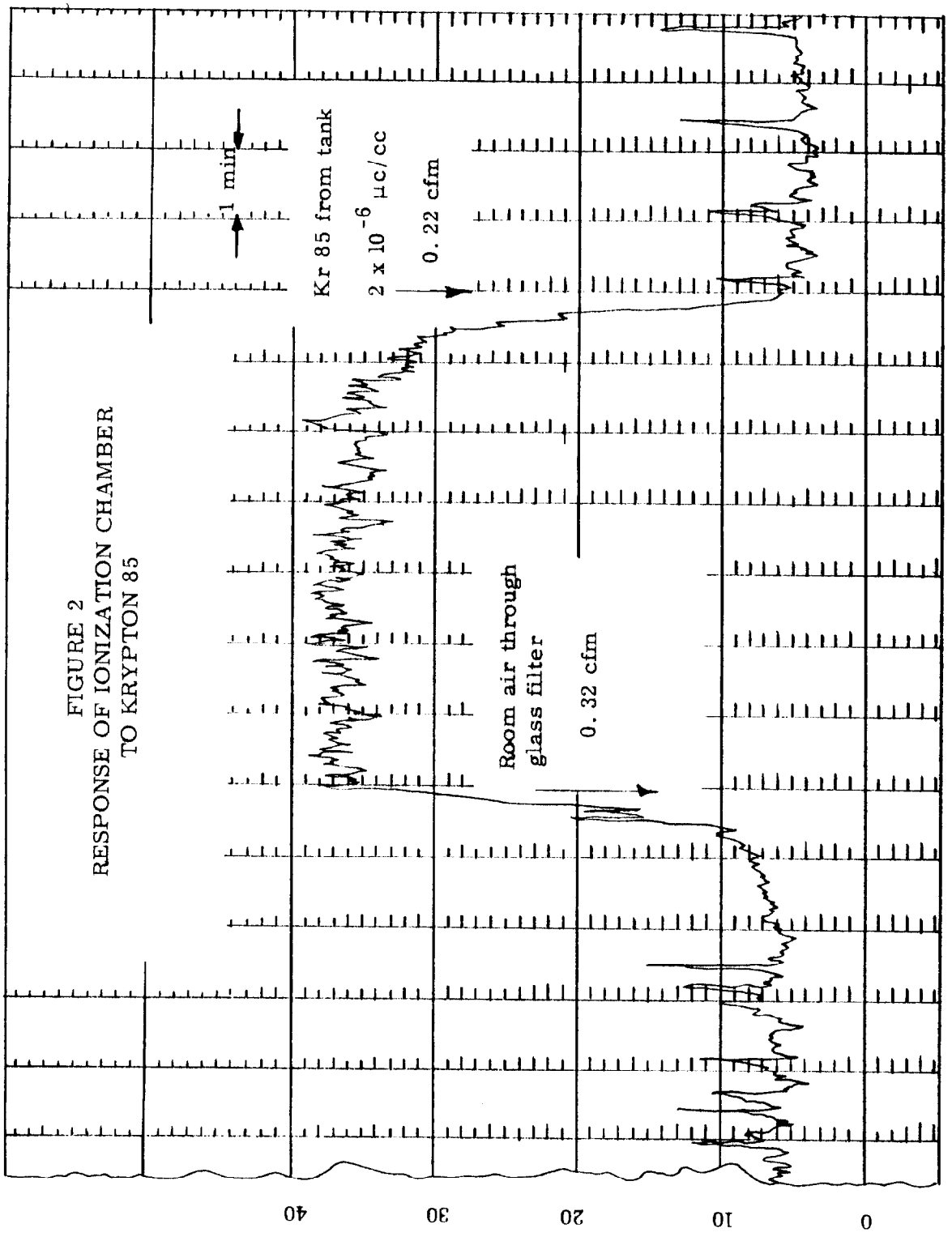


FIGURE 2
RESPONSE OF IONIZATION CHAMBER TO KRYPTON 85

Table 2 summarizes the data obtained in these experiments.

TABLE 2

RADIOACTIVITY RELEASES AS MEASURED BY THE 500 ml IONIZATION CHAMBER AND ELECTROMETER

Date	Time of Release	Duration of Release (Minutes)	Peak Electrometer Reading Amperes x 10^{15}	Background	
				Before Release (amperes x 10^{15})	After Release (amperes x 10^{15})
Aug. 2, 1963	8:20 A.M.	160	11.4	3.3	3.9
9, 1963	8:20 A.M.	15	11.1	6.0	6.3
9, 1963	1:20 P.M.	10	8.4	6.0	6.0
13, 1963	8:15 A.M.	15	9.0	4.5	4.5
15, 1963	8:25 A.M.	5	12.0	5.4	5.4
16, 1963	8:15 A.M.	20	11.1	4.2	4.2
22, 1963	7:55 A.M.	420	12.6	3.3	3.3
23, 1963	8:10 A.M.	5	10.8	3.3	3.3
24, 1963	8:05 A.M.	3	8.4	3.0	3.0
26, 1963	8:10 A.M.	5	8.4	3.6	3.6
27, 1963	8:10 A.M.	5	8.1	3.6	3.6
28, 1963	8:15 A.M.	15	12.0	3.3	3.3
29, 1963	8:10 A.M.	10	8.4	3.0	3.0

CONCLUSIONS AND RECOMMENDATIONS

The lowest gas concentration detectable is not the sole criterion for selection of a gas detection method. Simplicity, reliability, availability and cost are also factors which must be considered in the ultimate choice of a system. Information on reliability of equipment and of results is needed and must be obtained from laboratory studies and from information on field performance.

Additional operational data should be obtained for the different systems mentioned in this report - the cylindrical and pancake thin window counters; spherical ion chambers; large Kanne chambers; solid state detectors; and scintillation detectors. Commercial micromicroammeters should be tested to determine the lowest currents which can be measured reliably under field conditions.

In Table 3 the lowest levels of Krypton 85 which can be monitored under different operating conditions are shown. It is seen that for levels of the

order of 10^{-7} microcuries/cc, a simple cylindrical G-M counter is adequate if a large volume can be viewed, and it is the simplest detector available. If for reasons of shielding or space the sample volume must be small, an ionization chamber can be used. Alternately, the gas can be compressed into a small volume and viewed with a scintillation crystal or solid state detector. To achieve maximum sensitivity, the background must be reduced as much as possible. Scintillation or solid state detectors which view the gas under pressure and are shielded and covered with an umbrella of detectors in anticoincidence to minimize the response to cosmic radiation, should give the highest sensitivities. Although additional experimental data is needed to give the lowest levels detectable, these levels are probably of the order of 10^{-8} c/cc for a fifty per cent change in the background.

TABLE 3

LOWEST CONCENTRATIONS OF KRYPTON-85 WHICH CAN BE MONITORED BY VARIOUS DETECTORS

Detector	Level μc/cc
2.8 liter ion chamber, current reading	2.5×10^{-7}
2.8 liter ion chamber, rate of charge	1.3×10^{-7}
Kanne chamber, current reading	2.0×10^{-7}
Pressurized ion chamber, current reading	0.6×10^{-7}
Pressurized ion chamber, rate of charge	0.3×10^{-7}
Cylindrical G-M tube in infinite volume	0.5×10^{-7}
Solid state or thin scintillation detectors with cosmic-ray guard counters	0.1×10^{-7}

Calculations are based on an external radiation background of 0.02 mr/hr.

The MPC for occupational exposure to Kr-85, 40 hr week, is 10^{-5} μc/cc.

The MPC for continuous exposure of large populations is 3×10^{-7} μc/cc.

The sensitivity could also be increased, of course, by selectively concentrating Krypton before measurement, but this would involve much more complicated equipment and is outside the scope of this paper. (14)

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A RADIOACTIVE WASTE INCINERATOR

by

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Abstract:

A 50 lb/hr incinerator for the concentration of low-level radioactive combustible waste has been designed and fabricated under joint sponsorship of the U. S. Atomic Energy Commission and the U. S. Army. The experimental facility utilizes auxiliary gas firing to maintain high combustion gas temperatures of 2,000 to 2,500°F, and dry collection methods for gas cleaning. An evaluation program is presently being initiated at the U. S. Army Nuclear Defense Laboratory, Edgewood Arsenal, Maryland, where the facility is installed.

* * * * *

Introduction:

A 50 lb/hr incinerator for the concentration of low-level radioactive combustible waste has been designed and fabricated under joint sponsorship of the U. S. Atomic Energy Commission (AEC) and the U. S. Army. This experimental unit has been designed to investigate the effects of varying (1) the location, number, and orientation of overfire air inlets, (2) the inlet velocity and mass flow rate of primary and secondary combustion air, and (3) the use of auxiliary gas fuel to the primary and secondary combustion chambers. After optimum performance conditions are established, radioactive tracers will be utilized to evaluate the radiological limits of the unit, and actual radioactive waste will be incinerated. Upon satisfactory completion of the program, experimental equipment will be eliminated in a final prototype design. Gas cleaning equipment, which will be required to maintain control of the release of radioactivity within federal regulations, will consist of conventionally manufactured stock items.

Background:

The AEC has long been interested in combustion as a concentration and disposal technique for radioactive wastes. A review of incinerator designs and operational experiences was presented at the last Air Cleaning Conference[1] by R. Dennis and L. Silverman. Discontinued incineration projects at AEC installations resulted in an AEC-sponsored study of combustion by the U. S. Bureau of Mines. R. C. Corey, et al.,[2] concluded that (1) uniformity of combustion rate and maximum combustion efficiency for single-chambered incinerators designed for burning typical laboratory wastes (low bulk density, low ash, and relatively

high volatile content) was best achieved with an overfire air supply, and that (2) the mass flow rate and direction of overfire air could best be controlled by admitting air tangentially to a cylindrical combustion chamber.

At the completion of the Bureau of Mines study, the Harvard University Air Cleaning Laboratory (ACL), under AEC contract, studied the air cleaning problems associated with radioactive waste incineration. After conducting an extensive evaluation program on the Bureau of Mines unit, ACL proceeded to design and develop a complete incineration and air cleaning system. Experimental units, ACL I and ACL II, based on the Bureau of Mines design criteria for the combustion method, were built and evaluated. For the ACL II, a secondary combustion chamber and a new method of waste charging were added as improvements. This work was presented at the Sixth Air Cleaning Conference.[3]

The Department of the Army has assigned the responsibility for radioactive waste disposal to the U. S. Army Eastern Chemical Depot at Edgewood Arsenal, Maryland. One aspect of this responsibility is the disposal of large volumes of radioactive low-level combustible wastes including large animal carcasses. The U. S. Army Nuclear Defense Laboratory (USANDL), Edgewood Arsenal, Maryland, has been requested to study and recommend disposal methods for these wastes.

The common interest of the AEC and the Army has resulted in the present co-sponsored program under which the 50 lb/hr experimental incinerator has been designed and fabricated; this unit is about to be tested and evaluated. The design of this experimental facility was based on the evaluation of the ACL 25 lb/hr incinerator, and was developed by ACL with USANDL assistance.

A contract was awarded last year to Pyro Incinerator and Supply Corporation, Mineola, New York. The facility was to be installed at the U. S. Army Edgewood Arsenal by December 31, 1962, but because of many difficulties, installation is just now being completed. Initial studies, under the direction of USANDL and ACL, will include a detailed evaluation of the facilities performance. Since performance data are not presently available, we will devote this time to describing the facility and discussing our program.

Facility Requirements:

The following requirements have been established for the facility:

1. Burn dry combustible wastes, i.e. less than 10 percent moisture content, at a rate of not less than 60 lb/hr with auxiliary gas firing.
2. Burn wet combustible wastes, i.e. up to 60 percent moisture content, at not less than 40 lb/hr with auxiliary gas firing.
3. Safely burn combustible waste charges containing up to 200 μC/kg of radioactivity of any or all of the following materials; waste paper, towels, swab rags, surgical dressings, wood, animal carcasses, human or animal organs, rubber gloves, rubber or plastic tubing, and animal pen debris (sawdust, feces, vegetable matters).

Facility Description:

1. Incinerator: Figure 1 presents a flow diagram of the facility. The incinerator, on the left, consists of a cylindrical primary combustion chamber with a cylindrical secondary combustion chamber mounted directly above. Auxiliary gas firing will be provided in both chambers. Waste material will be introduced as packaged charges through the side charging box. The hot combustion gases exit tangentially from the top of the secondary combustion chamber to maintain the spiral gas flow pattern throughout the entire unit. The effluent gas is cooled by dilution with ambient air through the air dilution valve. The volume of cooling air required may vary from 1 to 4 times that of the total combustion air.

RADIOACTIVE WASTE INCINERATION FACILITY

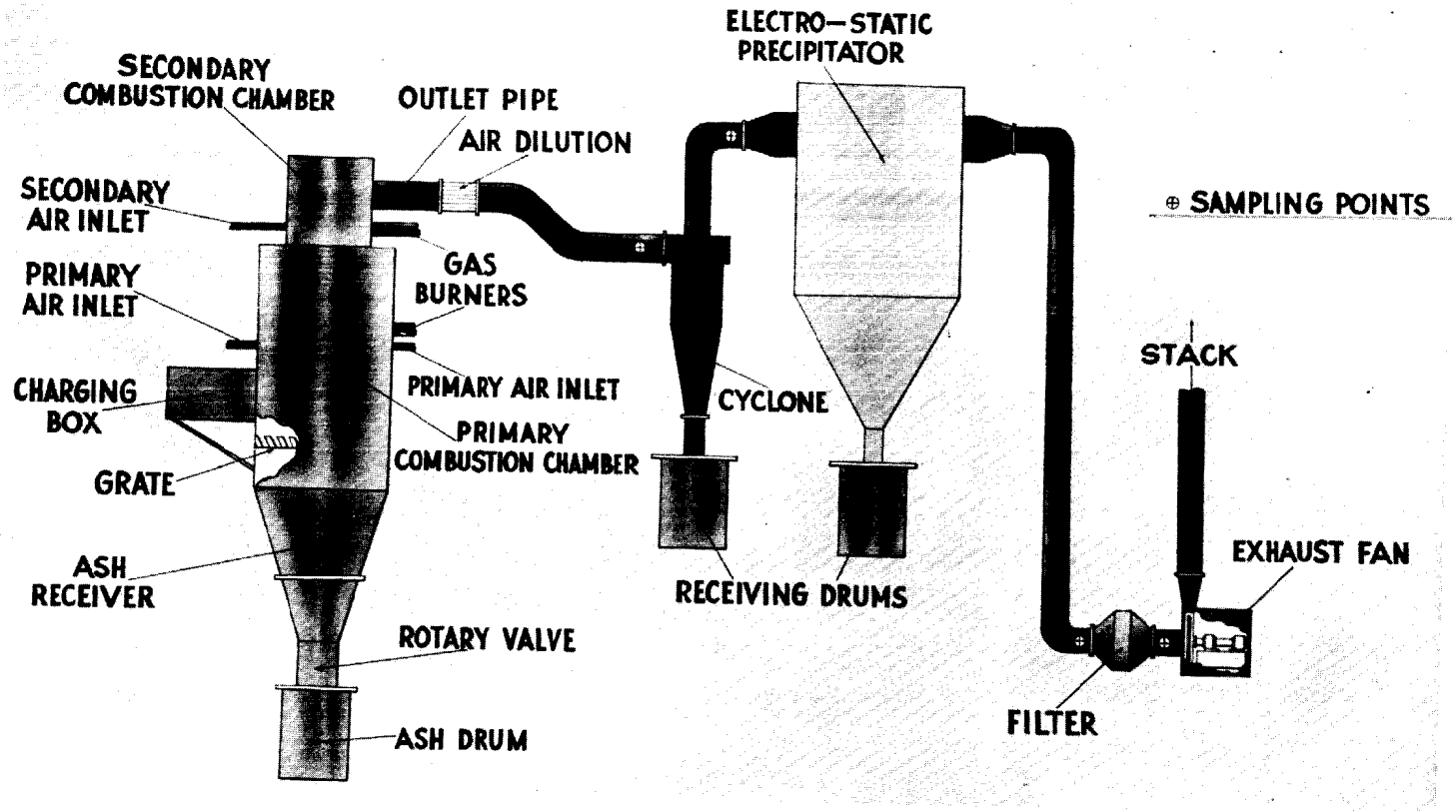

Figure 1

Since the incinerator is proposed for use in locations where liquid radioactive waste handling facilities are not available or are not desired, gas cleaning will be accomplished by dry collection methods; therefore, wet scrubbing techniques, which can cool as well as partially clean the gas, are excluded.

The system is composed of three basic units: (1) a medium velocity involute cyclone separator, which removes coarse particles entrained in the incinerator effluent and provides extended surface for gas temperature reduction, (2) a Cottrell-type electrostatic precipitator to collect fine solid particles, soot, mineral dust, and tar droplets formed by condensation of volatile organic vapors, and (3) a mineral fiber filtration unit, designed to capture any particulate blow-off from the electrostatic precipitator and to provide limited emergency protection in the event of electrical power failure or precipitator malfunction.

Figure 2 shows the facility lay-out as it is installed at the Edgewood Arsenal. Because of the space limitations, we have adapted a U-shaped configuration with the control panel placed in the center of the "U" so that the operator can observe most of the entire facility from the panel.

The incinerator shown in Figure 3 consists of a primary combustion chamber, 30 inches O.D. and 40 inches high, with a secondary combustion chamber, 20 inches O.D. and 32 3/8 inches high, mounted directly above the primary chamber. Both chambers are constructed from mild steel and lined with insulating refractory, 2 1/2 inches thick, to maintain a high combustion gas temperature of approximately 2,000 to 2,500°F.

Provision has been made to admit primary overfire air tangentially to the primary combustion chamber through two openings, approximately 24 inches above the top of the grate and 155° apart. The primary air inlets consist of 3-inch stainless steel pipes, into which specially shaped annular pipe sections, approximately 14 inches long, can be inserted to permit inlet velocity variation at constant flow rate. Two sets of air inlets, constructed as panel insert sections, are available. One pair is for tangential, horizontal air admission; the other for directing inlet air tangentially downward at a 30° angle relative to the horizontal. Total primary air, from either one or both inlets, is expected to range from 50 to 100 ft^3/min STP. Secondary combustion air, 20 to 40 ft^3/min STP, is admitted tangentially through a single 2-inch pipe, 6 1/2 inches above the base of the secondary chamber. Annular inserts are also provided to permit inlet velocity variation in the secondary air supply at constant flow rate.

Auxiliary gas firing will be provided by Eclipse Tunnel Mix Burners. Initially, it is planned to fire the secondary combustion chamber continuously to minimize discharge of unburned materials, and to fire the primary combustion chamber intermittently or continuously to maintain the desired operating temperature. Auxiliary gas firing will also be employed to preheat the combustion chamber to the desired operating temperature prior to charging waste. The primary burner is rated for 100,000 - 500,000 BTU/hr and the secondary, 50,000 - 250,000 BTU/hr. A platinum/platinum 10 percent rhodium thermocouple is located upstream of the dilution valve to measure the secondary chamber effluent gas temperature. The thermocouple is connected to a Bristol Free Vane Pyrometer Controller which regulates the Eclipse automatic control valve, thus providing automatic temperature control of the incinerator.

Waste material will be introduced as packaged charges, approximately 14 by 14 by 14 inches containing 6 to 10 pounds, through a side loading box, the bottom of which is located 9 inches above the grate. A three-pronged sliding fork inserted through the loading door permits temporary suspension of wet charges above the grate to facilitate rapid drying. A manually operated protec-

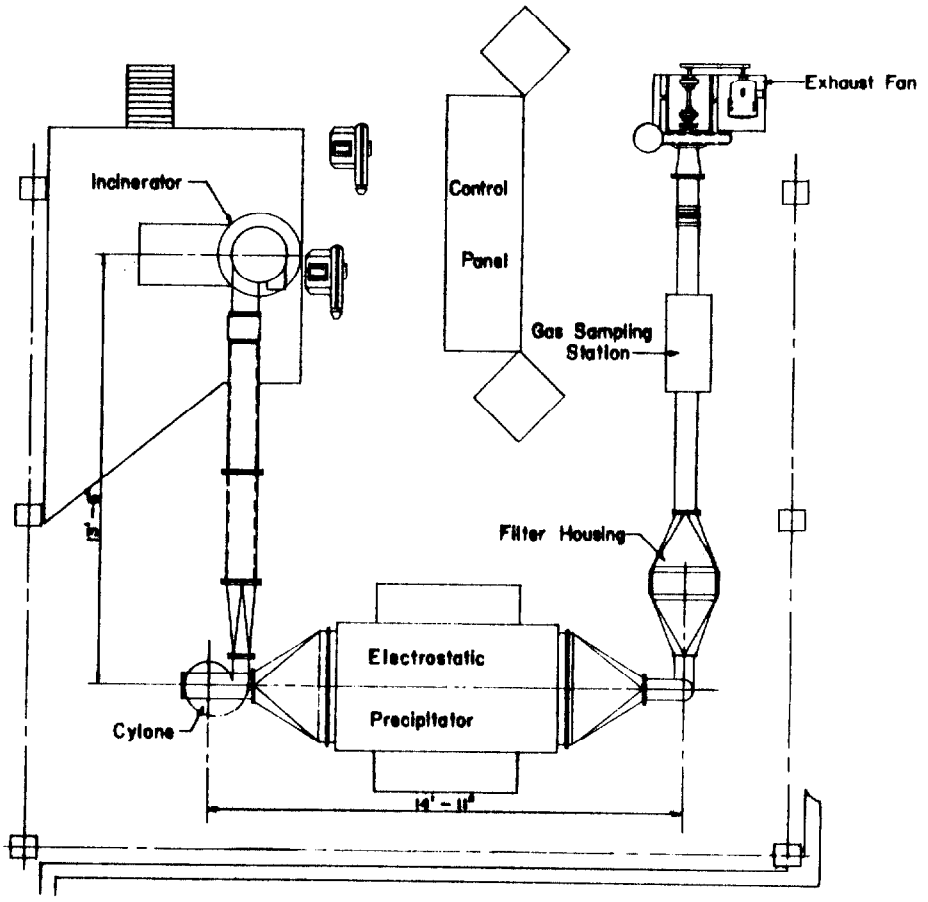

Figure 2

Incinerator Facility – Plan View

tive guillotine door, in conjunction with the totally enclosed charging box, is provided to minimize exposure of operating personnel to accidental release of dust or fumes during charging operations.

The incinerator grate, located 9 inches above the conical ash receiver, consists of four 40-pound cast-iron sections supported on a brick-work shoulder. The conical ash receiver flanged to the bottom of the combustion chamber discharges into a 55-gallon drum used for final collection of ash. A rotary valve with an air-tight shaft is attached to the bottom of the conical section to prevent ash spillage during change of ash drums. A hydraulic jack maintains a tight air seal between the drum rim and the incinerator hopper. Figure 4 shows a photograph of the incinerator during construction. The charge box and one air inlet panel have not been installed.

2. Gas Cleaning Equipment: The gas cleaning system, shown in Figure 5, is composed of a cyclone, electrostatic precipitator, and filter unit. The cyclone is capable of operating at gas flow rates to 760 ft^3/min STP and at temperatures to 750°F. Fractional collection efficiency at 70°F and 760 ft^3/min STP, shall be 95 percent for particles having a terminal settling velocity equivalent to those of 20-micron diameter spheres of specific gravity 2.0. The hopper discharge pipe is welded to a nonwarping steel cover plate and channel section. Gasketing, to be installed in the channel, will consist of 30 durometer Dow Silicon Rubber, 1/2-inch thick by 2 inches wide. A hydraulic jack will be used to maintain pressure contact between the 55-gallon steel drum and the gasket to prevent air leakage.

The cyclone effluent enters the 30 kV dc rod and plate electrostatic precipitator. It will furnish an average weight collection efficiency of 98 percent at gas flow rates to 1000 ft^3/min and at temperatures to 600°F. It is anticipated that submicron particles will be collected by operating the oversized unit at a decreased flow rate, and by not shaking the unit during operation. The precipitator will be cleaned by shaking methods when the incinerator is not in use and the material will be collected in the ash receiver located below.

The gas then enters the filter unit, which consists of two high-efficiency fiber filters in a parallel arrangement; MSA Dustfoe Space Filters rated for flow rates up to 1,000 ft^3/min STP will be used and will operate at air temperatures up to 500°F. The filter holder consists of two sixteen-gauge galvanized transition sections expanding from an 8-inch duct flange to a rectangular opening of approximately 2 by 4 ft. The filter holder is designed to handle commercial type filters of 24- by 24-inch face area and contains a clamping device so that units ranging in depths from 4 to 12 inches may be utilized. The filter holder is clamped between the transition sections with bolts and nuts around the exterior of the unit.

3. Control Instrumentation: Figure 6 shows the left end of the control panel. The facility is completely controlled from the control area and all blowers, electrostatic precipitator, etc., are connected by interlocks so that the auxiliary gas burners in the incinerator cannot fire unless the main exhaust fan is running. The interlock also prevents the shaker mechanisms on the electrostatic precipitator from operating while the main exhaust fan is operating. A key bypass is available for component testing. The timer system is utilized to control sampling times. The four sampling pumps can be started simultaneously and then turned off manually, or automatically by the timer. A continuous wind velocity and direction recorder is installed with the generator unit being placed at the same height as that of the top of the exhaust stack. The manometers indicate flow rates and pressure drops throughout the system. Figure 7 shows the remainder of the manometers and the second rack containing the Bristol

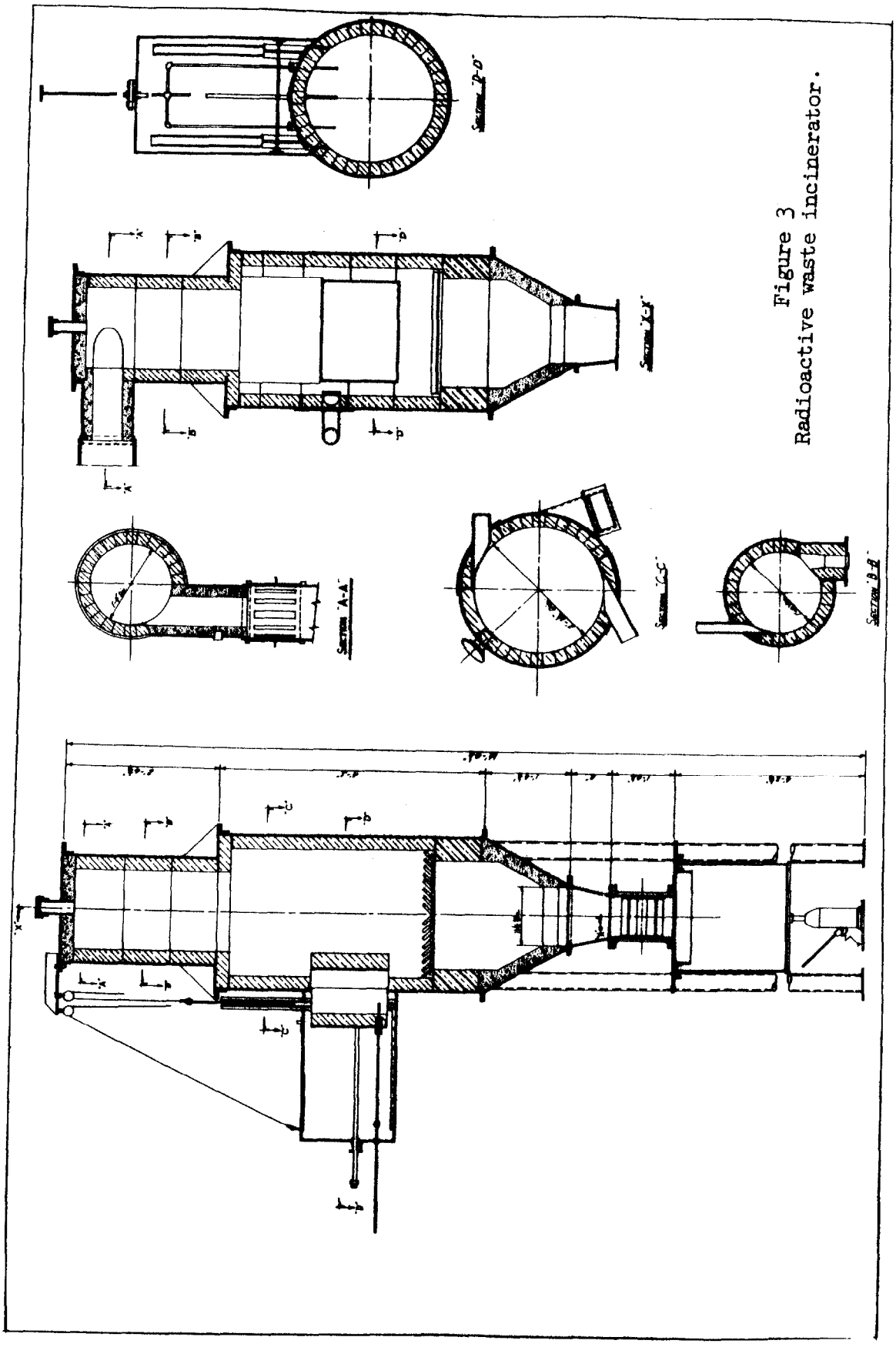

Figure 3
Radioactive waste incinerator.

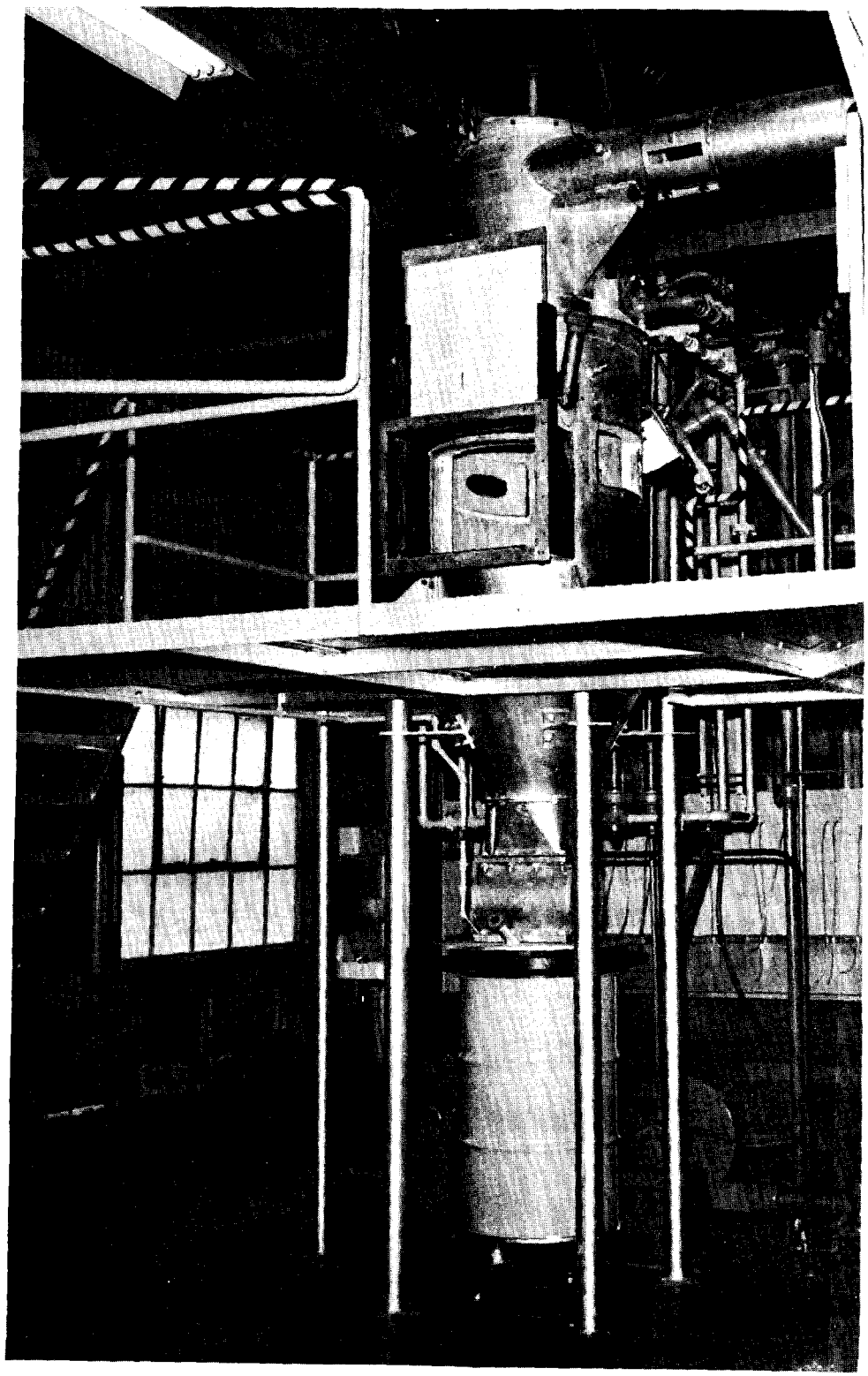

Figure 4
Radioactive waste incinerator.

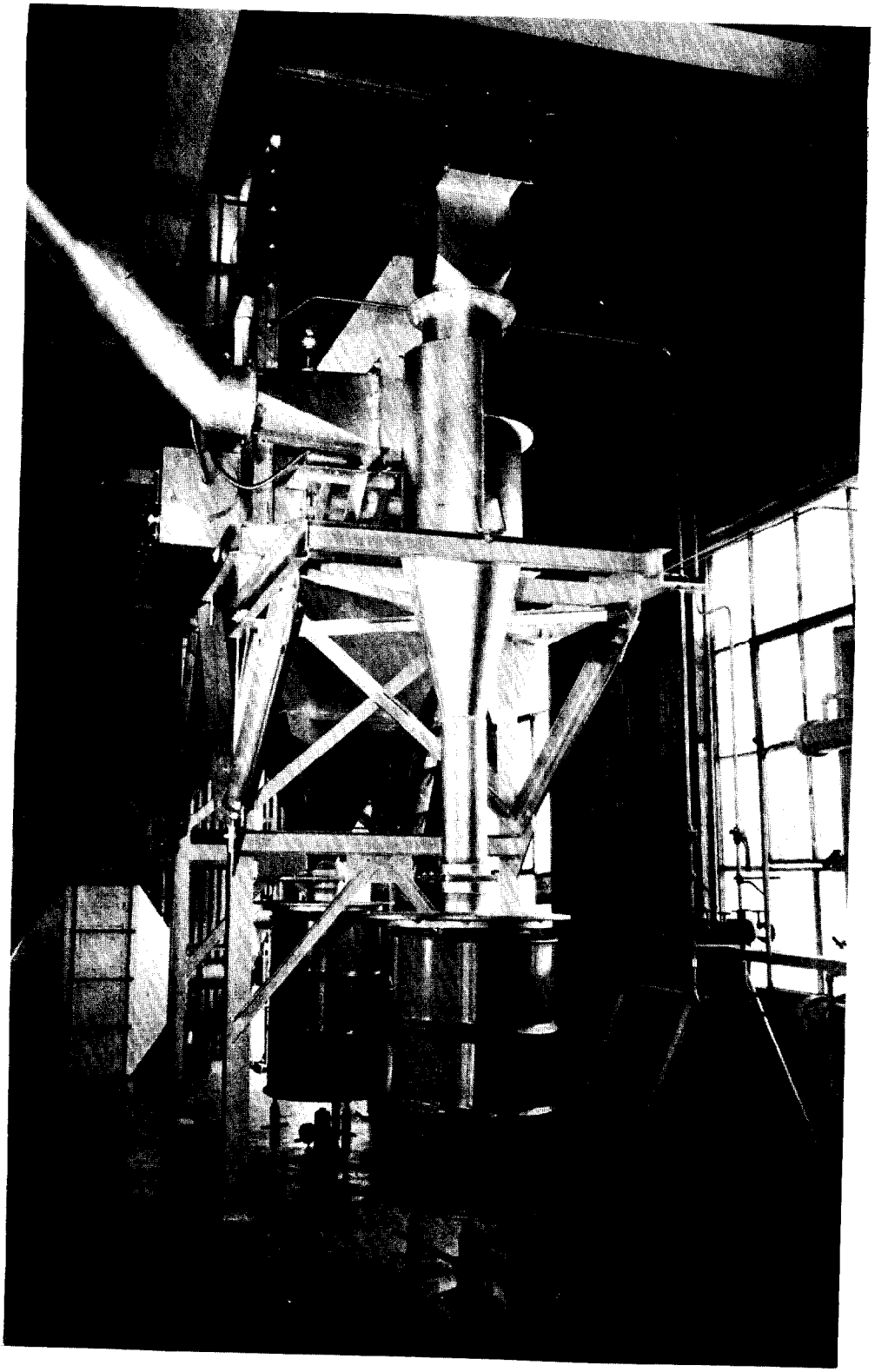

Figure 5
Gas cleaning system for radioactive waste incinerator.

Figure 6
Left end of control panel for a radioactive waste incinerator.

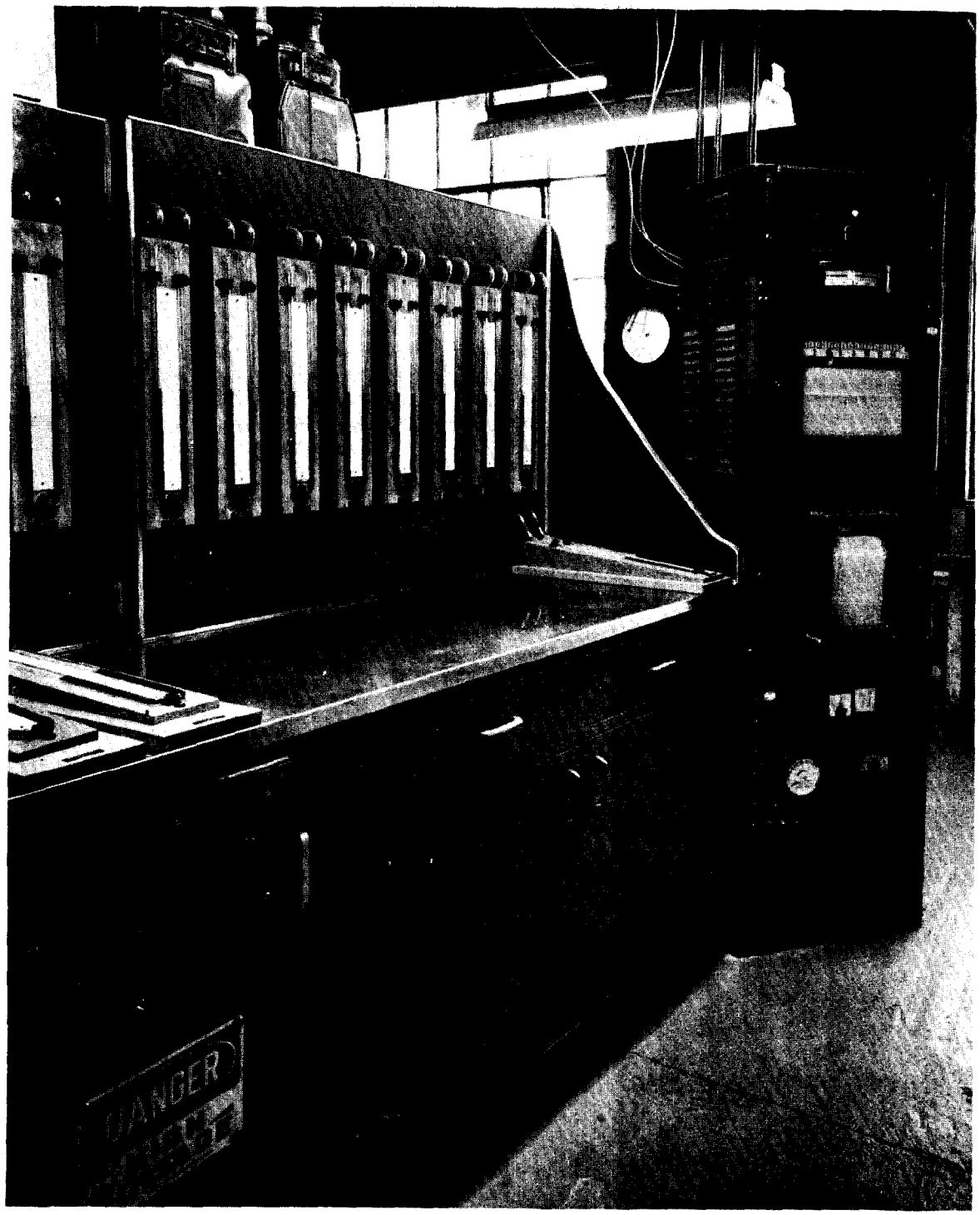

Figure 7
Right end of control panel for a radioactive waste incinerator.

indicator-controller for the incinerator. A multipoint recorder that records the temperatures at midbrick locations within the incinerator plus the temperature at each of the four sampling stations is located below the Bristol unit. Additional channels are available for experimental purposes. Finally, a continuous air monitor recorder is located at the bottom. The sampler for the air monitor is located near the charging box, which we feel to be the most hazardous position.

Figure 8 shows an isokinetic duct sampling station which includes a thermocouple, sampling probe and Pitot-static tube. The Pitot-static tube is located on the rear of the duct. In addition this is the gas sampling station. During sampling, a minimum of 100 bulb volumes is flushed through one of the gas sampling bulbs. When the sample has been taken, the valves are adjusted to switch the flow to the second bulb, allowing the first bulb to be removed from the system. Samples of the gas are then analyzed by a Fisher Gas Partitioner for carbon dioxide, oxygen, and nitrogen. The other three sampling stations consist only of the sample probe, thermocouple, and Pitot-static tube.

Program:

After the initial acceptance tests, which are presented in detail in NDL report[4] TM-8, the radiological treatment evaluation will involve a completely "cold" or nonradioactive engineering parameter study in which optimum conditions for efficient incineration and gas collection will be determined. This will be accomplished by studying such independent variables as: (1) air flow rate through the primary and secondary combustion chamber air inlets, (2) the air entry angle of the above mentioned inlets [0° or 30° relative to horizontal], (3) the rate of gas flow required for auxiliary burning, (4) time required to burn a given weight charge, (5) composition and temperature of combustion products, and (6) temperature distribution within the incinerator and about the grate.

Heat input, heat balance, and material balance will be evaluated under steady-state conditions by determining: (1) approximate analysis of the charge composition, to obtain a gross heating value; (2) temperature, humidity, and dew point, to obtain combustion air conditions; and (3) gas and proportional air rates, to obtain the total heating value of the auxiliary fuel.

In all tests of this phase, initial charges will not be radioactive and will consist of sawdust, typical waste, typical waste plus carcasses, and carcasses, in this order. Upon defining the experimental limits, radioactive materials will then be utilized to complete this phase of the evaluation.

Particulate samples will be collected before and after the cyclone, the electrostatic precipitator, and the filter units. These samples will be utilized to determine; (1) the dust loading of the gas stream, (2) the cleaning efficiency of each unit and, (3) the radiological decontamination factors.

To determine the operating efficiency and maximum permissible radioactive charge to the incinerator, specific nuclides will be employed. The isotopes, strontium 89, radium 226, and iodine 131, have been chosen because of their hazard and wide usage by the AEC and the Army. It is tentatively planned to introduce controlled amounts of these isotopes into carefully weighed and sealed packages of sawdust. Sampling of the effluent gas stream will permit evaluation of each air cleaning operation for each nuclide. Activity concentration will then be increased to determine the maximum amount of each isotope that can be handled safely with the facility. The system is designed for particulate removal and is not expected to effectively remove gases or vapors such as iodine, phosphorus, sulfur or carbon.

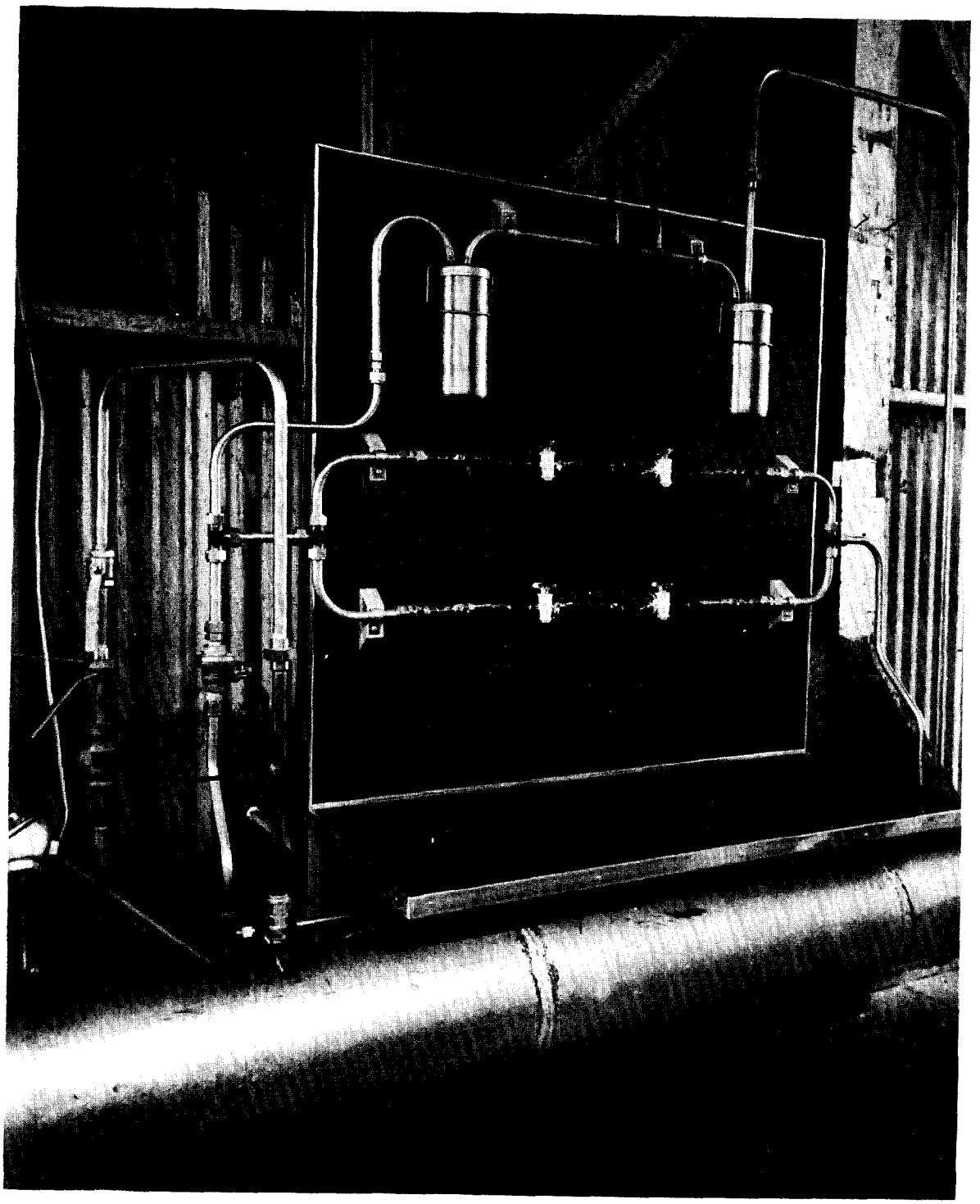

Figure 8
Isokinetic dust and gas sampling station for a radioactive waste incinerator.

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DISCUSSION AND COMMENT

In the reactor containment following maximum credible accident, one will expect to have some rather intense, local heat forces which would tend to superheat the air-water vapor mixture locally. These sources might coincide also with the fission products. Now, comment on the stability of an aqueous foam in the vicinity of the heat source.

We would hope that one could use sequential treatment, but in trying to meet that objective we did release steam at the same time we released foam, and we also tried to use the heated source for this, but we could not get the equivalent heat conditions.

Actually, if you consider reactor accident of the loss of coolant type, you have steam first, followed by the fission products. So the question is when do you use the foam, immediately, or as soon as your steam has been released?

I don't think the heat source that is present in an accident would be the kind that would disturb me. The reactor vessel, and all the rest, are insulated for the operating temperature, and you are not going to get above operating temperatures. Maybe I am wrong on that thinking, but after all, your reactors, your coolant is at, say, five or six hundred degrees, and at 2,000 psi, and then in the accident you release all this steam at that pressure, and get the equivalent energy release. The stored energy is rather tremendous in a water reactor, as you know. But whether there would be any source hotter than the reactor vessel, I don't know. The shielding, of course, has to take care of that.

I don't visualize a very large, hot ball situation; I visualize steam as our fundamental problem, and we tried steam releases, and again as a condenser foam is able to compete with this, but only on the basis of the same equivalent heat sink value.

The foams we have investigated are effective as fire-fighting media. The foams differ only in the sense that we add a reactant to them, like sodium thiosulfate, but the original idea of this type of foam generator goes to the Sheffield Mine Research Establishment, in England, where this was tried for fighting mine fires. The intent was that you could use it more as a blockage mechanism to keep oxygen from getting to the fire, and you would generate foam of this type. They used simply a brattice cloth into which was sprayed a detergent solution, and there would be air flow through the mine drift to actually form the foam.

As far as actually fighting fires go, the unit which you may want to see later today is one that they have at Idaho at the NRTS for firefighting. There are several more of them around the country. And I would rather let some safety fire expert, talk about the fire-protection qualities of the foam.

The optimum generation of Hi-Ex foam for fire extinguishment presupposed some opening in the structure to permit the air to escape.

Our concept is that we treat the air within the containment in a recirculating fashion. We just run the air through the unit, and fill the chamber up, and as you might note, one of the things is a question of whether

or not you plug the supply line to the blower. By having this line near the top of the vessel, all that is left is a small mount, and in the tests that we have run, we do not get the foam to flow down the small intake pipe. There is too much resistance, it just draws air from above the foam column.

So, what we are really doing, in its simplest form, is treating a reservoir of air, changing it into a reservoir with millions of capsules inside of aqueous foams.

We thought of other foams than aqueous foams. We are working with porous concrete. But we don't think anybody will let us do this in their containment vessel.

Richland Operations Comment: My comment is a comment, and not a question, on the fire-fighting capabilities. Actually, high-expansion foam seems to have tremendous potential in specialized uses in fire-fighting problems, and facilities of handling and using radioactive material, not as a replacement for sprinkler systems, or some of the other conventional methods, but because of its unique application in areas where personnel are excluded and some unusual approaches are necessary.

One of the phenomena of this is the fact that high-expansion foam has a difficult time extinguishing a candle, for instance, In fact sometimes you can't extinguish a single candle burning, because you have just enough oxygen in each of those little bubbles to keep the flame going. If you have a raging gasoline fire of, say, 5,000 gallons of gasoline at one time, it will do a beautiful job in a matter of two or three minutes. We had a rather extensive series of demonstrations at Hanford in May, and I know that Idaho had some in January.

The foam can also be frozen and it will stand outside in a bank.

One characteristic of the foam is very desirable; you may have noticed someone walking through it. You could actually crawl through it, or walk through it if this room is filled to the top, and although you could get a bit disorientated on your balance, you have no problem breathing, and only very slight irritation to the eyes. There is no better protection for people who are caught in one of these kinds of an incident; fire, explosion, whatever it might be than to dive in the foam, stay in it, and work your way out.

We burned an old building at Hanford. We introduced the foam by an indirect method, and it had to make two 90-degree turns to get into the space, fill the space, and control the fire in the glove box. After the glove box gloves had burned out, the foam flowed through the opening and extinguished the fire.

I was in the room while this was going on, to give more of an eye-witness review of it.

I had to find my way through, and find out where the heat was. It is possible to get within six inches of metal that was probably at a temperature of 1500 degrees, and not feel the heat. So, foam has great thermal protection for people who are caught in situations.